C. Van der Heyden

Operation and characterization of air scrubbers for the emission reduction of ammonia, hydrogen sulphide and greenhouse gases from animal housing systems

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Operation and characterization of air scrubbers for the emission reduction of ammonia, hydrogen sulphide and greenhouse gases from animal housing systems

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Dutch translation of the title:

Werking en karakterisering van luchtwassers voor de emissiereductie van ammoniak, waterstofsulfide en broeikasgassen vanuit stalsystemen.

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Cover: Picture of the structured packing material covered with biofilm inside the biological air scrubber at ILVO.

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Ghent, December 2017

Caroline Van der Heyden

Drench yourself in words unspoken

Live your life with arms wide open

Today is where your book begins

The rest is still unwritten

Table of Contents

List of Abbreviations

abbreviation	description
AOB	Ammonia Oxidizing Bacteria
BC	Background Concentration
bp	base-pair
BREF	Best Available Techniques (BAT) reference document
CA	Concentration Analysis
CSTR	Continuous Stirred-Tank Reactors
DF	Driving Force
EBRT	Empty Bed Residence Time
EC	Electrical Conductivity
EU	European Union
FA	Free Ammonia
FI	Flow Indicator
FNA	Free Nitrous Acid
GC	Gas Chromatography
L/G	Liquid-to-Gas
LI	Level Indicator
LR	Loading Rate
MOB	Methane Oxidising Bacteria
NEC	National Emission Ceilings
nMDS	non-Metric Multidimensional Scaling
NOB	Nitrite Oxidizing Bacteria
OTU	Operational Taxonomic Unit
PM	Particulate Matter
PR	Production

RE	Removal Efficiency
RR	Removal Rate
RSD	Relative Standard Deviation
SD	Standard Deviation
TAN	Total Ammonia Nitrogen
VOC	Volatile Organic Compounds
WWTP	Wastewater Treatment Plants

List of Symbols

abbreviation	characterization	unit
ε	void fraction of the packing	-
λ	thermal conductivity	$W.m^{-1}.K^{-1}$
τ	retention factor	-
μ	dynamic viscosity	Pa.s
μ_{max}	maximum specific growth rate	d^{-1}
ρ	volumetric concentration, volumetric mass	$kg.m^{-3}$
ΔH_V	evaporation enthalpy	$J.kg^{-1}$
a	specific surface area	$m^2.m^{-3}$
$A_{LW} = w_p \cdot l_p$	cross-sectional area	m^2
b	specific decay rate	d^{-1}
cp_G	packing specific coefficient for the gas phase	-
cp_L	packing specific coefficient for the liquid phase	-
c_p	specific heat	$J.kg^{-1}.K^{-1}$
C	concentration	$g.m^{-3}$
D	diffusion constant	$m^2.s^{-1}$
d_h	hydraulic diameter of the packing	m
DF	driving force of ammonia mass transfer	$gN.\ m^{-3}$
d_p	particle diameter	m
$E_{act}^{nitritation}$	activation energy of nitritation	$kJ.mol^{-1}$
$E_{act}^{nitratation}$	activation energy of nitratation	$kJ.mol^{-1}$
f	fraction	-
g	acceleration of gravity	$m.s^{-2}$

abbreviation	characterization	unit
h	heat transfer coefficient	$W.m^{-2}.K^{-1}$
h_L	liquid hold-up of the packing	$m^3.m^{-3}$
h_p	height of the packing	m
H	henry coefficient (gas to water)	-
i_{NXB}	nitrogen fraction in particulate inert components	$g\ N.(g\ COD)^{-1}$
k_G	gas mass transfer coefficient	$m.s^{-1}$
k_L	liquid mass transfer coefficient	$m.s^{-1}$
K_L	overall mass transfer coefficient	$m.s^{-1}$
K_a	acidity constant	$mol.m^{-3}$
K_{NH}	affinity constant for total ammonium	$g\ TNH\text{-}N.m^{-3}$
K_{NO2}	affinity constant for nitrite	$g\ TNO_2\text{-}N.m^{-3}$
K_{NO3}	affinity constant for nitrate	$g\ N.m^{-3}$
K_{O2}	affinity constant for oxygen	$g\ O_2.m^{-3}$
l_p	length of the packing	m
M	molecular weight	$g.mol^{-1}$
nY	number of cells	-
P	pressure	Pa
Q	flow rate	$m^3.s^{-1}$
r_b	reaction rate of conversion	$gN.s^{-1}$
R	gas constant	$J.K^{-1}.mol^{-1}$
RE	removal efficiency	%
RH	relative humidity	%
RSD	relative standard deviation	%
SD	standard deviation	ppm
T	temperature	°C
u	velocity	$m.s^{-1}$
V	volume	m^3
w_p	width of the packing	m
X_{AOB}	biomass concentration AOB	$g\ COD.m^{-3}$
X_{NOB}	biomass concentration NOB	$g\ COD.m^{-3}$
Y	yield coefficient	$g\ COD.(g\ N)^{-1}$

Subscript	characterization
B	bacteria (AOB or NOB)
BT	buffer tank
cell	cell
D	discharge
EW	evaporated water
F	fluid (gas or liquid)
FW	fresh water
G	gas
int	interface
L	liquid
N	nitrogen
P	packing of the air scrubber
R	recirculation
st	steam
sv	saturated water vapour
v	water vapor
vent	ventilation

Superscript	characterization
i	cell number i
in	inlet
max	maximum allowed value or setpoint
min	minimum allowed value
out	outlet

Summary

Due to the intensification of the livestock sector to meet the growing global meat consumption, higher emissions of pollutants are released into the air. One of the most important is ammonia. Its release causes acidification and eutrophication, both of which lead to biodiversity loss. In 2010, 94% of the anthropogenic ammonia emissions in Europe were attributed to agriculture (UNECE/LRTAP, 2012), putting this sector for a large challenge. As a response, the BREF guideline and AEA guideline required large pig and poultry housing facilities to be ammonia emission-low. Besides ammonia, the exhaust air from these buildings also contains various odorous compounds and dust particles, leading to possible odour nuisance in nearby residential areas and public health risks. Animal building emissions also involve nitrous oxide and methane, which are strong greenhouse gases contributing to climate change. While air scrubbers were in the first place introduced to reduce ammonia emissions, they were soon also regarded as a valuable technique to reduce other pollutants such as odour and dust. While optimizing air scrubbers to meet new requirements concerning odour and particulate matter besides ammonia, it is therefore recommended to anticipate the growing attention towards greenhouse gases by investigating their possible removal and/or production in these systems.

Chapter 1 provides a **state-of-the-art** on the application of air scrubbers and biofilters as a mitigation technique at animal housing systems. The properties of the incoming exhaust air of pig and poultry housing facilities were summarized to get an overview of the disturbances to be dealt with. When confronting the legally required minimal removal efficiencies (especially for ammonia) with the performances of full-scale air scrubbers, it was clear that not all installed air scrubbers reach these requirements. The process design and control options for air scrubbers were subsequently assessed in detail to gain more knowledge on the underlying reasons and in view of possible optimization strategies.

Following the literature review, the **objectives of this doctoral research work** were formulated (**Chapter 2**). This work focuses on the design and operation of chemical, biological and multi-stage (combi) air scrubbers for the removal

of ammonia from pig housing facilities. Besides ammonia, attention was paid to emissions of the strong greenhouse gases nitrous oxide and methane and to emissions of hydrogen sulphide as a model odour component. The optimization of air scrubbers was investigated by combining modelling and simulation with experimental measurements at full-scale installations, as they work synergistically.

Even though the performance of air scrubbers can be highly varying over time (Chapter 1), it is in literature mostly evaluated based on point measurements. Therefore, in this work, both diurnal fluctuations and spatial variations were investigated during **intensive measurement campaigns at a chemical air scrubber, a biological air scrubber and two-stage biological air scrubber** installed at pig housing facilities in Flanders (**Chapter 3)**. Ammonia, nitrous oxide and methane at the inlet and outlet of the air scrubbers were monitored on-line for one week using a photoacoustic gas monitor. Only one of the three air scrubbers performed well for ammonia removal. Nitrous oxide was produced inside the biological and two-stage scrubber and methane could not be removed in the different air scrubbers because of its low water solubility.

Modelling and simulation are useful tools to investigate the influence of design parameters and control handles on process performance in a time-efficient and cost-effective way. A **mechanistic model for a countercurrent chemical air scrubber** was set up in **Chapter 4**. Mass balances for water, ammonia, hydrogen sulphide, nitrous oxide and methane were implemented. Besides, heat balances were introduced to estimate water consumption and asses the effect of temperature. Air scrubbers are exposed to highly changing inlet conditions in terms of ventilation rate, air temperature and pollutant load. The model allowed to study the separate influence of each of these variables, which are interrelated in practice. The ventilation rate was found to have the largest influence on the ammonia removal efficiency in chemical air scrubbers.

A **mechanistic model of a biological air scrubber** was set up and applied (**Chapter 5**) to investigate in detail the role of the biological conversions in the removal efficiency. It was found that the effect of nitrification on the driving force for ammonia mass transfer mainly lies in the associated pH decrease,

rather than in the ammonia conversion as such. Without pH control, only about 50% of the absorbed ammonia is oxidized to nitrite and nitrate, balancing the pH increase from ammonia absorption by the protons produced during nitrification. Complete conversion of ammonia was possible when the pH was controlled at around 7, maintaining the driving force for mass transfer and consequently the ammonia removal efficiency.

The results of a long-term online monitoring campaign performed at two biological air scrubbers for one year, are presented in **Chapter 6.** In addition to continuous monitoring of the gas phase **concentrations of ammonia and nitrous oxide**, the most important **washing water characteristics** were analysed each week as well: pH, electrical conductivity (EC) as a measure for the total nitrogen content, as well as total ammoniacal nitrogen (TAN), nitrite and nitrate. Doing so, the air scrubber's performance could be linked with the washing water characteristics, thus increasing process knowledge for both the start-up period and during full operation. Additionally, the **influence of inoculation with activated sludge** was investigated as possible cheap and easy method to increase the performance of biological air scrubbers. The inoculated air scrubbers, immediately showed nitrifying activity, thereby reducing the pH and thus increasing the driving force. Furthermore, less nitrite accumulation was observed.

During the start-up of these two biological air scrubbers, the **microbial community structure** was investigated as well by applying metabarcoding (**Chapter 7**). Analysis of different samples of the inoculum, biofilm and washing water over five months showed that both biological air scrubber evolved to a rather comparable community structure, compared to the activated sludge. Small differences, mostly in abundancy, were observed between both air scrubbers and between the different stages. Nitrite oxidizing bacteria were found only abundant in the inoculated air scrubber, corresponding with the lower nitrite accumulation found in this system as seen in Chapter 6.

Chapter 8 offers some final considerations, conclusions and perspectives on the design and operation of chemical, biological and multi-stage air scrubbers, including suggestions for future research.

Samenvatting

De veeteeltsector in Vlaanderen heeft de laatste decennia een schaalvergroting ondergaan, wat met zich meebrengt dat de impact van de individuele bedrijven op hun omgeving potentieel groter wordt. Een van de belangrijkste emissies uit de veehouderij is ammoniak, verantwoordelijk voor verzuring en eutrofiëring, die beide leiden tot verlies aan biodiversiteit. In 2010 kon 94% van de antropogene ammoniakemissies in Europa toegeschreven worden aan de landbouw (UNECE/LRTAP, 2012), waardoor deze sector een grote uitdaging had. De wetgeving antwoordde hierop met onder andere de BREF en AEA-richtlijnen, die verplichten om grote varkens- en pluimveestallen ammoniak-emissiearm (AEA) te bouwen. Naast ammoniak bevat de uitlaatlucht van deze gebouwen ook verschillende geurcomponenten en stofdeeltjes, wat leidt tot mogelijke geuroverlast in nabijgelegen woongebieden en volksgezondheidsrisico's. De uitgaande gassen van stallen bevatten ook lachgas (distikstofmonoxide) en methaan, sterke broeikasgassen die bijdragen tot klimaatverandering. Terwijl luchtwassers in de eerste plaats werden ingevoerd om de ammoniakemissies te verminderen, werden ze spoedig ook beschouwd als een waardevolle techniek om andere verontreinigende stoffen zoals geur en stof te verminderen. Bij het optimaliseren van luchtwassers om aan al deze nieuwe eisen te voldoen, wordt daarom aanbevolen om op de groeiende aandacht voor broeikasgassen te anticiperen en hun mogelijke verwijdering en/of productie in deze systemen te onderzoeken.

Hoofdstuk 1 geeft achtergrondinformatie over de **huidige stand van zaken** van luchtwassers en biofilters als een mitigatietechniek bij dierlijke huisvestingssystemen. De eigenschappen van de inkomende uitlaatlucht van de stallen voor varkens en pluimvee werden samengevat, om een overzicht te krijgen van de te behandelen lucht. Bij het vergelijken van de minimale wettelijk vereiste verwijderingsefficiënties (vooral voor ammoniak) met de prestaties van volleschaal luchtwassers, werd het duidelijk dat niet alle geïnstalleerde luchtwassers aan deze eisen voldoen. Het procesontwerp en de controleopties voor luchtwassers werden vervolgens gedetailleerd beoordeeld

om meer kennis te verwerven over de onderliggende oorzaken en mogelijke optimalisatiestrategieën.

Na de literatuurstudie werden de doelstellingen van dit doctoraal onderzoekswerk geformuleerd (**Hoofdstuk 2**). Dit werk richt zich op het ontwerp en de sturing van chemische, biologische en meertraps (combi) luchtwassers voor het verwijderen van ammoniak (NH_3) uit varkensstallen. Naast ammoniak werd aandacht besteed aan de uitstoot van de sterke broeikasgassen lachgas (N_2O) en methaan (CH_4) en aan emissies van waterstofsulfide (H_2S) als modelgeurcomponent. De optimalisatie van luchtwassers werd onderzocht aan de hand van modellering en simulatie in combinatie met experimentele metingen op volleschaalinstallaties.

Hoewel de prestaties van luchtwassers sterk kunnen variëren in de loop van de tijd (Hoofdstuk 1), worden ze meestal geëvalueerd op basis van puntmetingen. Daarom werden in dit werk zowel dagschommelingen als ruimtelijke variaties onderzocht tijdens **intensieve meetcampagnes bij een chemische, een biologische en een tweetraps biologische luchtwasser**, geïnstalleerd in varkensstallen in Vlaanderen (**Hoofdstuk 3**). Ammoniak, lachgas en methaan werden aan de inlaat en uitlaat van de luchtwassers gedurende een week online gemeten met een foto-akoestische gasmonitor. Slechts één van de drie luchtwassers haalde de verreiste ammoniakverwijderingsefficiëntie van 70%. Lachgas werd geproduceerd in de biologische luchtwassers en methaan kon in geen enkele luchtwasser worden verwijderd door zijn lage wateroplosbaarheid.

Modellering en simulatie zijn handige hulpmiddelen om de invloed van ontwerpparameters en stuurvariabelen op procesprestaties op een effectieve en kostenefficiënte manier te onderzoeken. In **Hoofdstuk 4** werd een **mechanistisch model voor een tegenstroom chemische luchtwasser** gepresenteerd. Massabalansen voor water, ammoniak, waterstofsulfide, lachgas en methaan werden geïmplementeerd. Bovendien werden warmtebalansen geïntroduceerd om het waterverbruik in te schatten en het effect van de temperatuur te beoordelen. Luchtwassers worden blootgesteld aan sterk veranderende inlaatomstandigheden in termen van ventilatiedebiet, luchttemperatuur en belasting van polluenten. Het model liet toe de

afzonderlijke invloed van elk van deze variabelen, die in de praktijk met elkaar verbonden zijn, te bestuderen. Het ventilatiedebiet bleek de grootste invloed te hebben op de ammoniakverwijderingsefficiëntie in chemische luchtwassers.

Een **mechanistisch model van een biologische luchtwasser** werd opgesteld en toegepast om de rol van de biologische conversies op de verwijderingsefficiëntie in detail te onderzoeken (**Hoofdstuk 5**). Het effect van nitrificatie op de drijvende kracht voor ammoniakmassaoverdracht werd onderzocht. Daarbij werd aangetoond dat het grootste effect hoofdzakelijk ligt in de bijbehorende pH-afname, in plaats van in de ammoniakomzetting als zodanig. Zonder pH-regeling wordt slechts ongeveer 50% van de geabsorbeerde ammoniak geoxideerd naar nitriet en nitraat, door de balans tussen de pH-verhoging bij ammoniakabsorptie en de productie van protonen tijdens nitrificatie. Volledige omzetting van ammoniak was mogelijk als de pH werd geregeld op ongeveer 7, waarbij de drijvende kracht voor massa-overdracht werd behouden.

Hoofdstuk 6 rapporteert de resultaten van de online monitoringcampagne op lange termijn (één jaar) op twee biologische luchtwassers. Naast de continue monitoring van de **concentraties aan ammoniak en lachgas in de gasfase**, werden de belangrijkste **waswatervariabelen** elke week geanalyseerd: pH, elektrische geleidbaarheid (EC) als maat voor het totale stikstofgehalte, evenals totale ammoniakale stikstof (TAN), nitriet en nitraat. Hierdoor konden de prestaties van de luchtwassers gekoppeld worden aan de waswatervariabelen, waardoor proceskennis werd opgedaan, zowel tijdens de opstartperiode als tijdens de volledige werking. Daarnaast werd de **invloed van inoculatie met actief slib** onderzocht als mogelijke gemakkelijke en goedkope methode om de prestaties van biologische luchtwassers te verhogen. De geïnoculeerde luchtwasser vertoonde onmiddellijk nitrificatieactiviteit, waardoor de pH daalde en daarmee de drijvende kracht verhoogde. Bovendien werd minder nitriet-accumulatie waargenomen.

Tijdens de opstart van deze twee biologische luchtwassers werd eveneens de **microbiële gemeenschapsstructuur** onderzocht door metabarcoding toe te passen (**Hoofdstuk 7**). Analyse van verschillende stalen van inoculum, biofilm

en waswater over vijf maanden liet zien dat in biologische luchtwassers de microbiologie zich ontwikkelt tot een vrij vergelijkbare gemeenschap in vergelijking met het actief slib. Kleine verschillen, meestal in hoeveelheid, werden waargenomen tussen beide luchtwassers en tussen de verschillende trappen van de luchtwassers. Nitrietoxiderende bacteriën werden alleen gedetecteerd in de geïnoculeerde luchtwasser. Dit komt overeen met de lagere nitrietaccumulatie die in deze luchtwasser werd gevonden, zoals in Hoofdstuk 6 werd aangetoond.

Ten slot biedt **Hoofdstuk 8** conclusies over het ontwerp en de werking van chemische, biologische en meertrapsluchtwassers, inclusief een aantal overwegingen en suggesties voor toekomstig onderzoek.

1

Air scrubbers and biofilters:

state-of-the-art

1.1 Abstract

The global intensification of livestock production resulted in potentially higher emissions of ammonia, odour, particulate matter (PM) and greenhouse gases (nitrous oxide and methane). Air scrubbers were introduced as a low ammonia emission housing technique. However, regulations with regard to the use of air scrubbers changed, including required removal efficiencies for odour and PM besides ammonia. In practice, however, the required removal efficiencies for these pollutants are not always obtained, indicating the need of process optimization in terms of process design and/or operation. While optimizing air scrubbers, it is argued and recommended to anticipate the growing attention towards greenhouse gases, such as methane and nitrous oxide, which are present in exhaust air from animal housing facilities. However up till now, very little is known about the behavior of greenhouse gases in air scrubbers. Moreover, the formation of nitrous oxide in (biological) air scrubbing systems cannot be excluded. This contribution summarizes the state-of-the-art of air scrubbers for the reduction of emissions of ammonia, odour, nitrous oxide, methane and fine dust and points out perspectives for process optimization in terms of design and control. The air and liquid flow configuration, packing dimensions and packing material should be carefully considered. Control options for water flow rate, water discharge and acid dosage need to be optimized. Dosage of apolar solvents and inoculation of the packing material can be innovative control options to achieve a better removal of less water-soluble components.

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1.2 Introduction

The global livestock sector has intensified significantly during the last few decades to meet the growing meat consumption (FAO, 2006, 2013), resulting in potentially higher concentrations of pollutants to be released in the air. The most regulated pollutant in agriculture is ammonia. Its release in the atmosphere causes acidification, its presence in surface water causes eutrophication, both of which lead to biodiversity loss (Oenema, Velthof, Amann, Klimont, & Winiwarter, 2012). To minimize the pressure on the environment, a multi-pollutant protocol was agreed upon in 1999 by the EU, the United States and Canada under the Convention on Long-Range Transboundary Air Pollution, i.e. the so-called Gothenburg protocol. Besides, Europe implemented the National Emission Ceilings (NEC) directive (2001/81/EC), which imposed on each Member State maximum emission levels to be reached by 2010 for four pollutants related to acidification, eutrophication and ground-level ozone production, namely sulphuric oxide, nitrogen oxide, volatile organic compounds (VOC) and ammonia. A total ammonia reduction of 17% was to be reached by 2010 compared to 1990 (Gothenburg protocol, 1999), a goal which was reached by 23 of the 27 EU member states (EEA, 2013). The Gothenburg protocol was revised in 2012, aiming for a 6% reduction of ammonia in 2020 compared to 2005 (Gothenburg protocol, 2012). On 31 December 2016, a revision of the NEC directive entered into force, also including fine particulate matter as 5th pollutant to cut health impacts by air pollution. All member States must transpose the Directive into national legislation by 30 June 2018 and produce a National Air Pollution Control Programme by 2019 setting out measures to ensure that emissions of the five main air pollutants are reduced by the percentages agreed by 2020 and 2030 (European Commission, 2016).

As 94% of the European ammonia emissions is due to agriculture (UNECE/LRTAP, 2012), this sector has a great effort to make in achieving the Gothenburg protocol and NEC directive. Livestock rearing accounts for 64% of the global anthropogenic ammonia. Given that most of the global emissions from livestock production (31-55%) originate from animal housing and storage

systems (Beusen, Bouwman, Heuberger, Van Drecht, & Van Der Hoek, 2008), the Gothenburg Protocol advised to implement emission-low animal housing facilities. In the European Union (EU), the focus lies on pig and poultry housing facilities, which have a large contribution in the ammonia emission. Several source-based and end-of-pipe techniques are applied for ammonia emission reduction. Reduction at the source can be established through manure collection surfaces which are easy to clean, size reduction or cooling of the emitting surfaces or by quickly removing manure from the animal housing facility, keeping the retention time of manure in the animal house low (BREF, 2003). End-of-pipe solutions comprise chemical, biological or combined air scrubbers and biofilters, in which the exhaust air is led through a wet packed bed to remove water soluble components. They are widely applied in pig housing facilities in several countries (Jacobsen, 2011; Lemay et al., 2009; Melse & Ogink, 2005). In the Netherlands, the share of fattening pigs and sows housed in emission low housing systems increased from 50% in 2010 to more than 60% in 2012, an increase which can almost fully be attributed to air scrubbers (CBS, 2012).

Besides ammonia, also the release of odorous components may cause local nuisance, especially in densely populated areas (CBS, PBL, 2012; Platteau, Van Gijseghem, Van Bogaert, & Maertens, 2012; Radon et al., 2004). Because of their limited spatial impact, specific regulations on odour emission are typically imposed at a local, regional or national level (Melse, Ogink, & Rulkens, 2009). In the case of persistent odour related complaints, odour reduction techniques like air scrubbers or biofilters are required.

The emission of dust or particulate matter (PM) attracts a lot of attention as well, as it can enter the respiratory system of both humans and animals (Wathes et al., 1998). It is mostly classified according to size in two main categories, namely PM_{10} and $PM_{2.5}$, defined as particulate matter which passes through a size-selective inlet with a 50% efficiency cut-off at 10 and 2.5 µm aerodynamic diameter, respectively (1999/30/EC). Several types of pollutants can be adsorbed to dust particles. A major concern in this respect is the formation of bio-aerosols through the association of micro-organisms with dust particles

(Cambra-López, Aarnink, Zhao, Calvet, & Torres, 2010; Seedorf et al., 1998). Also gases and endotoxins, antibiotics, insect parts, pollen and grain particles can interact with particulate matter (Millner, 2009). A recent study in the Netherlands, using 57.000 health reports of nearby residents of farms, showed both positive and negative effects (RIVM, 2016). People living nearby farms have less asthma and allergises and less COPD (chronical long disease), but for people who are already vulnerable, a higher risk was found. It is unclear whether the extra instances of pneumonia in the studied area are caused by specific pathogens that originate from animals (zoonotic agents), or by people becoming more susceptible to pneumonia through exposure to substances emitted by livestock farms, such as particulate matter, endotoxins and ammonia (RIVM, 2016). Livestock production of the EU represents 8% of the total PM_{10} emissions and 4% of the total primary $PM_{2.5}$ emissions (Klimont et al., 2008). The Gothenburg protocol 2012 includes a 22% reduction of $PM_{2.5}$ emissions between 2005 and 2020. Air scrubbers can reduce PM emission from animal housing facilities as the dust is partly captured in the washing water. Required reduction efficiencies are taken up in legislation in several European countries (Melse et al., 2009).

Greenhouse gases are another important category of pollutants related to the livestock sector, having a share of 14.5% in the global anthropogenic greenhouse gas emissions (Gerber et al., 2013). The pig and poultry sector contribute 9 and 8% to the sector's emissions, respectively. Methane and nitrous oxide are two strong greenhouse gases, having a global warming potential of 34 and 298 CO_2 equivalents, respectively (IPCC, 2013). The emission of methane, nitrous oxide and carbon dioxide accounts for 44%, 29% and 27% of the total livestock sector's greenhouse gas emissions, respectively. The Kyoto protocol, signed in 1997, at the United Nations Framework Convention on Climate Change (UNFCCC), put forward a global reduction of greenhouse gas emissions by 5.2% on average by 2012 compared to 1990 (UN, 1998). Several parties also adopted an amendment to the Kyoto protocol to reduce greenhouse gas emissions by at least 18% below 1990 levels for the period 2013 to 2020 (UNFCCC, 2013). As agriculture and in particular the

livestock sector is a major contributor to greenhouse gas emissions, this sector can play an important role in their mitigation (Gerber et al., 2013). The reduction of nitrous oxide and methane emissions are important policy options concerning climate change mitigation (Brink, van Ierland, Hordijk, & Kroeze, 2005). While optimizing air scrubbers to meet new requirements concerning odour and particulate matter besides ammonia, it is therefore recommended to anticipate the growing attention towards greenhouse gases by investigating their possible removal and/or production in these systems.

This review summarizes the state-of the art and points out optimization perspectives for air scrubbers aiming at the reduction of ammonia, odour, PM and greenhouse gases. The general operating principle and different types of air scrubbers are discussed, pointing out the most important aspects regarding the design and operation of these installation. The properties of the incoming exhaust air of pig and poultry housing facilities are summarized to get an insight of the disturbances which must be dealt with. The legally required minimal removal efficiencies are confronted with the performances of full-scale air scrubbers in terms of the considered pollutants, also considering the influence of disturbance variables. Finally, the state-of-the-art concerning process design and process operation of air scrubbers is assessed in more detail.

1.3 General operating principle of air scrubbers and biofilters

Air scrubbers are mostly applied at mechanically ventilated housing facilities; their operating principle is illustrated in Figure 1.1. Air from the housing facility is withdrawn through fans towards a central duct and blown counter-current or cross-current into the air scrubber. In Flanders and the Netherlands, a pressure chamber of at least three meters length is required in front of the scrubber to equalize airflow and pollutant concentration (InfoMil, 2013). As under-dimensioning or implementing a by-pass is not allowed, air scrubbers have to be designed to cope with the total maximum ventilation capacity of the facility or peak pollutant load (InfoMil, 2013). Since 2015, chemical air scrubbers are also applied at non-mechanically ventilated cattle housing

facilities (Burgers, 2015). As the building is not completely closed, not all air is drawn through the air scrubber, but still high overall reductions (77% in the Netherlands) can be reached.

A washing liquid is distributed equally at the top of the scrubber. The packed bed must be wetted evenly (no dry spots) to avoid the escape of unwashed exhaust air. Intense contact between the gas and liquid phase ensures mass transfer of water soluble pollutants from the air (g) to the washing water (l). A chemical equilibrium is established, e.g. between ammonia (NH_3) and ammonium (NH_4^+):

$$NH_{3(g)} + H_2O_{(l)} \leftrightarrow NH_{3(l)} + H_2O_{(l)} \leftrightarrow NH_{4(l)}^{+} + OH_{(l)}^{-} \qquad \textbf{Eq.1.1}$$

The washing water is recirculated to reduce water consumption. Eventually the concentration of accumulated contaminants in the washing liquid becomes too high, such that discharge of the washing water is necessary. To maintain operational stability of the system, constant measurements are necessary with a pH- and electrical conductivity (EC)-electrode in the buffer tank or recirculation line, controlling the dosage of acid (if applicable) and discharge of washing water. Fresh water is added to compensate for the water loss due to discharge and evaporation, which can be controlled with a level sensor in the buffer tank. A demister behind the scrubber prevents the escape of small droplets from the air scrubber.

In a chemical air scrubber for ammonia removal an acid is added to the washing water to keep the pH between 1.5 and 4, shifting the equilibrium towards ammonium and thereby increasing the absorption capacity. Sulphuric acid (H_2SO_4) is typically used (Eq.1.2). Deposition of the formed ammonium sulphate ($(NH_4)_2SO_4$) takes place when the maximum solubility of the salt (772 $g.L^{-1}$ or 164 $gN.L^{-1}$ at 20°C; Weast, 1975) is exceeded, resulting in clogging and thus a higher energy requirement. To prevent this, in Flanders and the Netherlands the content of ammonium sulphate in the washing liquid is not allowed to exceed 2.1 $mol.L^{-1}$, or 58.8 g $N.L^{-1}$, as a safety requirement which is about 3 times lower than the maximum solubility (InfoMil, 2013;

MB19/03/2004). A demister behind the scrubber is mandatory for chemical air scrubbers to prevent acid droplets from entering the environment.

$$2NH_3 + H_2SO_4 \rightarrow (NH_4)_2SO_4 \qquad \textbf{Eq.1.2}$$

In biological air scrubbers for ammonia removal, ammonium captured in the washing water is slowly oxidized to nitrite (NO_2^-; Eq.1.3) and subsequently to nitrate (NO_3^-; Eq.1.4). This process is called nitrification.

$$NH_4^+ + 1.5O_2 \rightarrow NO_2^- + 2H^+ + H_2O \qquad \textbf{Eq.1.3}$$

$$NO_2^- + 0.5O_2 \rightarrow NO_3^- \qquad \textbf{Eq.1.4}$$

These conversions are carried out by ammonia oxidizing bacteria (AOB) such as Nitrosomonas and by nitrite oxidizing bacteria (NOB) such as Nitrobacter and Nitrospira respectively (Kristiansen, Pedersen, et al., 2011). These microorganisms are suspended in the washing liquid or attached to a biofilm on the packing material. Two possible configurations of biological air scrubbers are encountered in practice. A biotrickling filter, as illustrated in Figure 1.1, contains microorganisms immobilized in a biofilm on the packing material. This type is mostly used at animal housing facilities as the installation cost is less compared to the second type, a bioscrubber, which consists out of an absorption tower and a separate bioreactor (VDI 3478 Part 1, 2011). The latter reactor contains the microorganisms in suspended growth, can be aerated and is mostly inoculated with activated sludge from wastewater treatment plants (WWTP) (Delhoménie & Heitz, 2005). Besides, the growth of a biofilm cannot be excluded in these systems, but the abundancy of the bacteria is not necessarily the same (Sercu, Núñez, Van Langenhove, Aroca, & Verstraete, 2005). The higher the liquid flow rate applied, the higher the shear stress on the biofilm, which can strongly influence biofilm erosion and sloughing. The start-up of biological air scrubbers may take several weeks or even months (Ottosen et al., 2011). Water is periodically discharged from a biological air scrubber to prevent inhibition of AOB and NOB by free ammonia (FA) and free nitrous acid (FNA). In Flanders, the maximum allowed nitrogen content

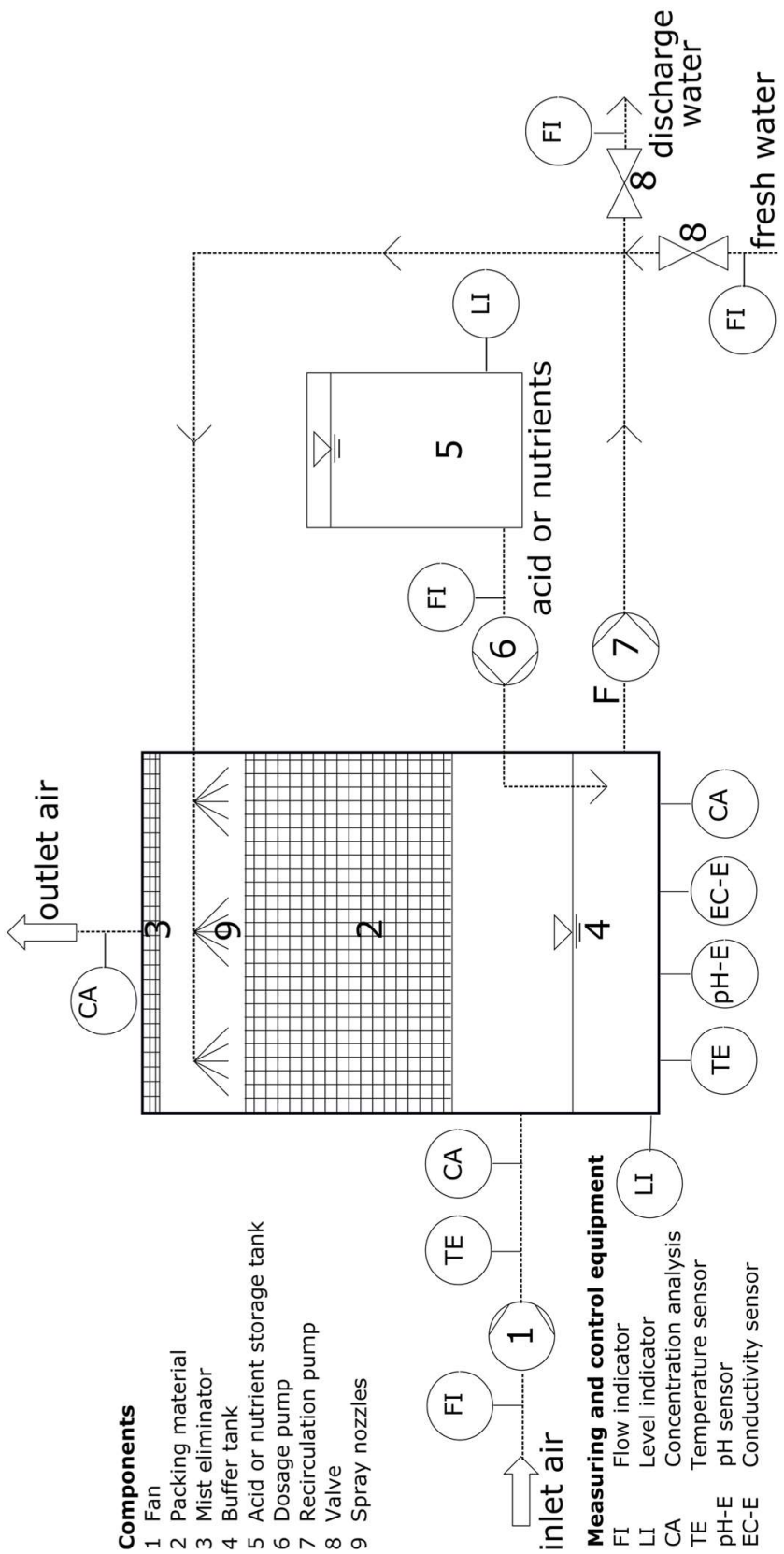

Figure 1.1. Simplified process diagram of a counter-current air scrubber. The storage tank of a chemical air scrubber contains acid. For a biological air scrubber a storage tank is optional and may contain necessary nutrients for the microorganisms.

of the washing liquid of a biological air scrubber is set at 3.2 $gN.L^{-1}$ (MB31/05/2011), to prevent inhibition of the bacteria. This implies a much larger amount of discharge water being produced compared to chemical air scrubbers. The pH must be kept within the favourable range of the microorganisms, typically between 6.5 and 7.5 (Ottosen et al., 2011).

Besides the nitrification reactions, a denitrification step may be included, involving the conversion of nitrate into nitrogen gas (Eq.1.5), reducing the nitrogen load in the washing water. Melse, Hol, & Nijeboer (2012) reported a reduction in the discharge water amount by a factor 3 by applying denitrification and still keeping the nitrogen content of the discharge water around 3.4 $gN.L^{-1}$.

$$NO_3^- \rightarrow NO_2^- \rightarrow NO + N_2O \rightarrow N_{2(g)} \qquad \textbf{Eq.1.5}$$

Biofilters constitute another biological system, consisting of a filter bed of organic material, which is kept humid but not necessarily subject to continuous water spraying. The same biological processes (Eq.1.3 and Eq.1.4) as in biological air scrubbers, can take place in biofilters. Nevertheless, they are not suitable for the treatment of exhaust air directly drawn from animal houses as they suffer from clogging and quick acidification by FNA accumulation due to the high dust and ammonia concentrations (Melse & Ogink, 2005). Recent legislation tends to no longer accept stand-alone biofilters as an ammonia emission reduction technique (BREF, 2017). However, they have been proven efficient for odour removal (Martens et al., 2001) and are mostly used in combination with a chemical or biological air scrubber.

When two or more scrubbing stages are placed behind each other, the overall system is termed a combined air scrubber. This type has evolved from the chemical and biological scrubbers for the combined removal of different types of pollutants, like ammonia, odour and dust. A chemical air scrubbing stage is very efficient for ammonia removal, while a biological air scrubbing stage or a biofilter are found more suitable for odour reduction (Aarnink, van Hattum, Hol, & Zhao, 2007; Hol, Groenestein, Dousma, Melse, & Ogink, 2010; Melse, Ogink, & Bosma, 2008). Water curtains may be introduced as a pre-treatment

to remove dust before the air enters the packing material, thus preventing clogging. Placing different stages behind each other also allows efficient reuse of the recirculation water. Washing water from a biological stage contains a low nitrogen content as the maximum allowed content is set at 3.2gN.L^{-1}. This implies that the water can still be used in the preceding chemical stage, increasing the nitrogen content further to the maximum allowed nitrogen content of 58.8 gN.L^{-1}. In this way, less fresh water addition is needed and the amount of discharge water is reduced compared to using separate washing water streams for each step.

Air scrubbers and biofilters need to be followed up regularly by the operator to check if they still function properly. The washing water parameters, i.e. pH, EC, discharge volume and electricity use need to be in between the required ranges (InfoMil, 2015; MB31/05/2011) and the spray pattern must be uniform, wetting the whole packing. These must give an indication if the air scrubber performs well, as measurements of ammonia removal efficiency are mostly not available. Implications are discussed more profoundly in Chapter 8.

1.4 Characteristics of exhaust air of pig and poultry housing facilities

The characteristics of exhaust air constitute the disturbance variables which should be taken into account while designing an air scrubber. These include ventilation rate, temperature, relative humidity, as well as the composition of the exhaust air. The ventilation rate in Flanders and the Netherlands ranges between 3 and 80 $m^3.h^{-1}.pig^{-1}$, and between 5.4 and 72 $m^3.h^{-1}.fowl^{-1}$, depending on the type and the weight of the animal (Klimaatplatform Pluimveehouderij, 2010; Klimaatplatform Varkenshouderij, 2014; Van Gansbeke, Van den Bogaert, & Vettenburg, 2009). At warm outside temperatures and high animal activity, a higher ventilation rate is needed to maintain the desired temperature in the building (CIGR, 1999) having a dilution effect on gaseous indoor concentrations (K. Y. Kim et al., 2007). The temperature of the exhaust air from piggeries lies between 18° and 26°C, the optimal temperature being 20 to 22°C (Banhazi, Seedorf, Rutley, & Pitchford, 2008; Geers et al., 1989). The

recommended temperature in poultry houses for rearing and broilers varies between the starting temperature of 32-34°C, with every week a decrease of 4°C to a minimum of 20°C (Hulzebosch, 2004). The relative humidity of the exhaust air depends on the outside temperature, the temperature of the animal housing facility, breathing of the animals, supply of drinking water and the manure and urine excretion (Van Gansbeke et al., 2009) and varies between 54 and 99% in pig houses (Radon et al., 2002). The ideal relative humidity for poultry is 60-80% (Hulzebosch, 2004).

Exhaust air from pig and poultry houses contains a variety of gaseous components and dust. The concentrations are influenced by the housing system, floor type, ventilation rate, animal age, etc. Table 1.1 gives an overview of the most common pollutants and odour components as well as the odour concentration itself and their concentration ranges found in pig and poultry housing facilities; conventional or emission-low. Background concentrations are given to allow comparison to the atmospheric values.

The indoor ammonia concentrations typically range from 5 to 20 ppm_v in well-ventilated animal housing facilities (Crook, Robertson, Travers Glass, Botheroyd, & Topping, 1991). However, sometimes concentrations higher than 50 ppm_v are measured, especially in poorly ventilated buildings (Radon et al., 2002; Ulens et al., 2014). The main source of ammonia is the hydrolysis of urea of urine by the faecal enzyme urease leading to ammonium formation in an aqueous medium, released as ammonia through volatilisation from the slurry pit or urine puddles (Cortus, Lemay, Barber, Hill, & Godbout, 2008). The atmospheric background concentration ranges between 0.7 and 20 ppb_v in Europe (EEA, 2014). Near some types of livestock areas, such as pigpens in the United States, local atmospheric concentrations have been reported to be as high as 47 ppm_v (ATSDR, 2004). The average global ammonia concentration in the atmosphere is 0.3–6 ppb_v (ATSDR, 2004).

Table 1.1. Literature overview of typical background concentrations and indoor pollutant and compound concentration ranges for pig and poultry housing systems. Odour components are classified in the four main groups.

Pollutant or component	BC(1)	Indoor concentration range at piggeries	Indoor concentration range at poultry buildings	Unit
NH_3 (N-containing)	0.7 – 2 [1]	2 [8] – 87 [11] [2; 3; 4; 5; 6; 7; 8; 9; 10; 11]	1 [13] – 50 [13] [4; 8; 12; 13]	ppm_v
Odour	<300 [14]	517 [15] – 18063 [15] [2; 5; 6; 15; 16]	278 [17] – 3032 [12] [12; 17]	$OU_E.m^{-3}$
H_2S (S-containing)		15 [18] – 6180 [8] [5; 8; 18; 19; 20]	30 [8] – 2240 [21] [8; 21]	ppb_v
Dimethyl sulphide (S-containing)		1.0 [22] – 9.8 [19] [19; 20; 22; 23]	0.25 [23] – 174 [24] [23; 24]	ppb_v
Acetic acid (fatty acid)		164 [22] – 386 [19] [19; 20; 22; 25]	4 – 1758 [24]	ppb_v
p-cresol (aromatic)		0.2 [23] – 25.7 [22] [19; 20; 22; 23; 25]	0.06 [23] – 14 [24] [23; 24]	ppb_v
Indole (N-containing)		0.05[19] – 0.7 [20] [19; 20; 22; 25]	0.4 [24] – 48 [24] [24]	ppb_v
CO_2	390.5 [26]	1000 [11] – 5000 [11] [2; 3; 9; 10; 11]	500[13] – 3000[13] [12; 13]	ppm_v
CH_4	1.803 [26]	7 [27] – 630 [11] [2; 3; 11; 27]	1 [13] – 10 [13] [12; 13; 21]	ppm_v
N_2O	0.324 [26]	0.28 [2] – 1.2 [11] [2; 3; 11]	0.20 [12] – 4 [13] [12; 13]	ppm_v
PM_{10}	3.0 – 172.5 [1]	150 [5] – 5500 [11] [2; 3; 5; 9; 10; 11]	101 [12] – 10360 [28] [12; 28; 29]	$\mu g.m^{-3}$
$PM_{2.5}$	1.5 – 52.4 [1]	27 [10] – 501 [9] [2; 3; 9; 10; 11]	31 [12] – 1140 [28] [12; 28; 29]	$\mu g.m^{-3}$
Total bacteria	$10 – 10^3$ [30]	$1{\cdot}10^5$ [31] – $2{\cdot}10^{10}$ [10] [10; 31; 32]	$3{\cdot}10^6$ [31] – $4{\cdot}10^{10}$ [32] [10; 31; 32]	$CFU.m^{-3}$
Total fungi	$10 – 10^3$ [30]	$5{\cdot}10^4$ [31] – $2{\cdot}10^5$ [18] [18; 31]	$1{\cdot}10^4$ [31]	$CFU.m^{-3}$

BC = Background concentration
* converted from $\mu g.m^{-3}$ to ppm or ppb

Reference: [1] EEA (2014)* [2] Melse, Hol, et al. (2012) [3] Van Ransbeeck (2013) [4] Groot Koerkamp et al. (1998) [5] Jacobson et al. (2006) [6] Melse & Mol (2004) [7] Melse & Ploegaert (2011) [8] Zhu et al. (2000) [9] Aarnink et al. (2007) [10] Zhao et al. (2011) [11] Ulens et al. (2014) [12] Melse, van Hattum et al. (2012) [13] Wathes et al. (1997) [14] Arends et al. (2008) [15] Van Langenhove & Defoer (2002) [16] Hove et al. (2012) [17] Mosquera et al. (2008) [18] Thorne et al. (2009) [19] Hansen, Adamsen, et al. (2012) [20] Jonassen et al. (2012) [21] Okoli et al. (2004)* [22] Feilberg et al. (2010) [23] Van Huffel et al. (2012)* [24] Trabue, Scoggin, et al. (2010)* [25] Trabue, Kerr, et al. (2010) [26] IPPC (2013); time span 2011 [27] Girard et al. (2009) [28] Takai et al. (1998) [29] Banhazi et al. (2008) [30] Jo & Seo (2005) [31] Seedorf et al. (1998) [32] Radon et al. (2002)

Odour components can be divided into four main groups: sulphur components, fatty acids, aromatics and N-containing components (Bottcher, 2001). Many different components can be present in the exhaust air, all with different odour

thresholds and different concentrations without all of them having been identified (Lunn & Van de Vyver, 1977; O'Neill & Phillips, 1992). The concentrations of odorous components are mostly found under 1 ppm_v (Hansen, Adamsen, et al., 2012). Odour concentrations are usually not measured chemically but with an olfactometer, which uses the human nose as a sensor (CEN EN 13725). The odour concentrations vary greatly among housing facilities and within individual housing facilities (Van Langenhove & Defoer, 2002). Different studies tried to identify the key odorants responsible for livestock odour (Gralapp, Powers, & Bundy, 2001; Hansen, Adamsen, et al., 2012; Hobbs, Misselbrook, & Pain, 1995; Trabue, Kerr, et al., 2010; S. Zhang et al., 2010). Many have been found but there is still no consensus about which odorants make up the complete olfactometric odour.

The concentrations of greenhouse gases methane and carbon dioxide vary widely, compared to the nitrous oxide concentration, which stays quite constant throughout the year (Van Ransbeeck, Van Langenhove, & Demeyer, 2013). Most of carbon dioxide can be attributed to animal respiration (Ni, Hendriks, Coenegrachts, & Vinckier, 1999), even though some is also produced in the manure pit (Ni, Vinckier, Hendriks, & Coenegrachts, 1999). Methane is produced by anaerobic digestion of organic matter by microorganisms, which occurs predominantly (65-70%) during manure storage (Girard et al., 2009). Nitrous oxide is a product of both nitrification and denitrification, and will be released at all livestock production stages where conditions are favourable for these processes. The highest emissions of N_2O from animal housing facilities in the UK, were noted from poultry housed on straw, as oxygen and carbon sources are fully available for nitrification and denitrification (Chadwick, Sneath, Phillips, & Pain, 1999).

Dust or PM in livestock housing facilities is almost entirely organic and originates from feed, skin and feathers, bedding and dried faeces (Carpenter, 1986). The levels of PM are the highest in broiler houses compared with other animal species, because of poultry litter (Cambra-López et al., 2010). The concentration range of bacteria and fungi are higher than the ones found in

residential buildings (Arya & Kaushik, 2013). However, their concentration as such is not a direct hygiene indicator, since this also depends on the species.

1.5 Performance of air scrubbers and biofilters

1.5.1 Regulatory requirements and guidelines

The regulatory requirements for air scrubber performance with regard to the reduction of pollutants, differ among countries (Table 1.2). In Flanders it is mandatory to reach at least 70% ammonia reduction during the whole year, independent of the used system (MB31/05/2011). In the Netherlands, Germany and Denmark, different types of air scrubbers are certified according to their removal efficiency for certain pollutants, which they need to meet upon installation (Danish EPA, 2007; VROM, 2007). An odour removal efficiency of 30%, as reported feasible by Ogink & Lens (2001), is recommended in Flanders and the Netherlands as the minimum removal efficiency for chemical air scrubbers (InfoMil, 2013; LNE, 2011). In Denmark, chemical air scrubbers are assumed not to reduce odour emissions (Danish EPA, 2015). The German government does not impose a certain odour removal efficiency but instead, the outgoing odour may not be observed as exhaust gas from animal housing facilities and may not exceed 300 OU.m-3 (Arends et al., 2008).

1.5.2 Overview of reported removal efficiencies

Table 1.3 gives an overview of the observed removal efficiencies for ammonia, odour, nitrous oxide, methane, PM_{10} and $PM_{2.5}$ for different types of full-scale air scrubbers in different countries. The empty bed residence time (EBRT) is defined as the empty bed volume of the packing material divided by the gas flow rate and reflects the available time for mass transfer.

1.5.2.1 *Ammonia*

Chemical air scrubbers can reach removal efficiencies for ammonia of up to 99%, even though a large variation is found and sometimes the required 70% removal is not reached (Table 1.3). When tested under controlled circumstances, the standard deviation on the removal efficiency can be kept as low as 1% (DLG test standards [ID8]). This shows the importance of good

Table 1.2. Reduction requirements and guidelines for different pollutants for four Western-European countries and regions

Air scrubber	Country/ Region	NH_3 (% red)	Odour (% red)	PM_{10} (% red)	$PM_{2.5}$ (% red)	Ref.
Chemical	The Netherlands	70/85/95	30/40	35/60	-	[1]
	Flanders	70	30	35	30	[2,3]
	Denmark	90	-	-	-	[4]
	Germany	70/95	-	70 [b]	-	[5]
Biological	The Netherlands	70/85	45	60/75	-	[1]
	Flanders	70	40	60[c]/75[d]	35[c]/75[d]	[2,3]
	Denmark	70	73	-	-	[4]
	Germany	70	<300 $OU.m^{-3}$, [a]	70[b]	-	[5]
Combined	The Netherlands	70/85	70/75/ 80/90	80	-	[1]
	Flanders	70	70	80[e]	70[e]	[2,3]
	Germany	70/95	<300 $OU.m^{-3}$, [a]	70[b]	-	[5]
Biofilter	The Netherlands	70	45	80	-	[1]
	Flanders	70	-	-	-	[2,3]
	Germany	-	<300 $OU.m^{-3}$, [a]	70[b]	-	[5]

a No crude gas odour may be observed
b Total dust content instead of PM_{10}
c Contact time < 2.0 s in the packing and wetted areas
d Contact time > 2.0 s in the packing and wetted areas
e Higher reductions are possible when they can be proved by a scientific measurement report of the system
[1] (VROM, 2013)
[2] (MB31/05/2011)
[3] (LNE, 2011)
[4] (Danish EPA, 2006)
[5] (Arends et al., 2008)

maintenance and follow-up. The ammonia removal efficiency of the biological air scrubbers discussed in Table 1.3 varies from 10% to 99%. Table 1.3 shows that a biofilter is not suitable for direct treatment of exhaust air because of its low ammonia removal efficiency (average of 15% [ID24] to 72% [ID21]); at some biofilters taken up in Table 1.3 ammonia was sometimes even produced (-9 [ID25] to -60% [ID22]) due to air leakage through the filter bed and/or ammonia stripping. These low removal efficiencies are mostly caused by an inadequate functioning of the humidification system, allowing the bypass of untreated air into the environment and increasing inhibition in the packing material as ammonium and nitrite accumulate (Melse & Hol, 2012). Increasing the moisture content from 20% to 50% improved the ammonia removal efficiency of biofilters over 30% (Hartung & Jungbluth, 2001; L. Yang, Kent, et al., 2014). Several different types of combined air scrubbers can be found in practice. Table 1.3 shows that the reported variation in ammonia removal efficiency for combined air scrubbers is smaller compared to the other types of scrubbers. A possible explanation is that the use of more stages provides more certainty to to remove more and multiple pollutants in case of some failure in one single stage. If multiple stages are used, the stages can be more specialised in removing a specific pollutant. A water curtain can be placed in front, to remove as much dust as possible. By this, the second stage will receive less dust, avoiding clogging. The second stage can then reduce the ammonia concentration further, as the water is cleaner than in the first stage, allowing a higher driving force of mass transfer. In the third stage, more odour can be removed, using for example a biofilter. The air is already moistened from the two previous stages and since less ammonia is present, the formation of nitrite/nitrate salts in the biofilter is avoided (Melse, Ogink, & Bosma, 2008). Reuse of the washing water of the biological section in the chemical section does not seem to have a negative effect on the ammonia removal efficiency (Melse et al., 2011).

1.5.2.2 *Odour*

The olfactometric measurement results are highly varying, ranging from negative removal efficiencies (odour production) to up to 80% efficiencies, for

Table 1.3. Reported pollutant removal efficiencies for different types of air scrubbers and biofilters for pig and poultry housing facilities

Type of air scrubber	**ID**	**Country Animals**	**EBRT (s)** range **μ ± σ**	**NH_3 (%)** range **μ ± σ**	**Odour (%)** range **μ ± σ**	**N_2O (%)** range **μ ± σ**	**CH_4 (%)** range **μ ± σ**	**PM_{10} (%)** range **μ ± σ**	**$PM_{2.5}$ (%)** range **μ ± σ**	**Reference**
Chemical	1	NL Pigs	>0.9	77 – 97 **91**	27 – 66	-	-	-	-	Vrielink et al. (1997); Melse & Ogink (2005)
Chemical	2	NL Poultry	-	40 – 99 **90**	-	-	-	-	-	Hol & Satter (1998); Melse & Ogink (2005)
Chemical	3	NL Pigs	>0.5	90 – 100 **99**	-	-	-	-	-	Verdoes & Zonderland (1999); Melse & Ogink (2005)
Chemical	4	NL Poultry	>0.4	76 – 100 **95**	-	-	-	-	-	Hol et al. (1999); Melse & Ogink (2005)
Chemical[(a)]	5	NL Poultry	>0.4	96 – 100 **98**	-	-	-	-	-	Wever & Groot Koerkamp (1999); Melse & Ogink (2005)
Chemical (90% ammonia)	6	NL Poultry	0.2 – 2	30 – 99 **77** ± *31*	-10 – 80 **48** ± *22*	-30 – 20 **1** ± *12*	-60 – 50 **-1** ± *25*	5 – 60 **33** ± *17*	0 – 55 **28** ± 22	Mosquera et al. (2011): Melse, Hofschreuder et al. (2012)
Chemical (70% ammonia)	7	NL Pi/Po	0.2 – 1.8	45 – 99 **76** ± *20*	-20 – 80 **19** ± *28*	-25 – 25 **-1** ± *12*	-90 – 20 **-5** ± *31*	20 – 65 **41** ± *20*	0 – 80 **33** ± *23*	Mosquera et al. (2011); Melse, Hofschreuder et al. (2012)
Chemical	8	DE Pigs	0.5 – 2.9	87 – 89 **88** ± *1*	-	-	-	81 – 99[(b)] **89** ± *8*	-	DLG5957 (2011)

Type of air scrubber	**ID**	**Country Animals**	**EBRT (s)** range **μ ± σ**	**NH_3 (%)** range **μ ± σ**	**Odour (%)** range **μ ± σ**	**N_2O (%)** range **μ ± σ**	**CH_4 (%)** range **μ ± σ**	**PM_{10} (%)** range **μ ± σ**	**$PM_{2.5}$ (%)** range **μ ± σ**	**Reference**
Chemical (pH<7) (biological)	9	NL Poultry	**0.56**	48 – 85 **69** ± *13*	-23 – 43 **17** ± *25*	-17 – 3 **-5** ± *7*	-9 – 12 **2** ± *8*	11 – 84 **44** ± *26*	13 – 46 **31** ± *17*	Melse, van Hattum, et al. (2012) Melse, Hofschreuder et al. (2012)
Biological (70% ammonia)	10	NL Pigs	0.4 – 1.8	50 – 85 **76** ± *16*	5 – 65 **42** ± *30*	-120 – -20 **-70** ± *42*	-5 – 10 **4** ± *7*	45 – 50 **48** ± *4*	10 – 50 **37** ± *16*	Mosquera et al. (2011); Melse, Hofschreuder et al. (2012)
Biological (70% ammonia)	11	NL Pi/Po	3.2 – 4.5	10 – 99 **59** ± *33*	-60 – 60 **-3** ± *49*	-400 – -5 **-208** ± *154*	-5 – 50 **10** ± *17*	45 – 90 **74** ± *13*	60 – 90 **75** ± *11*	Mosquera et al. (2011); Melse, Hofschreuder et al. (2012)
Biological	12	NL Pigs	**0.28**	46 – 92 **78** ± *18*	5 – 69 **42** ± *30*	-120 – -21 **-70** ± *42*	-6 – 10 **4** ± *7*	-	-	Melse & Ploegaert (2011)
Biological	13	NL Pigs	**0.54**	41 – 92 **79** ± *17*	-29 – 87 **49** ± *30*	-	-	-	-	Melse & Mol (2004)
Biological	14	NL Pigs	>0.34	42 – 92 **82**	-	**-78**	-	-	-	Melse, Ploegaert, et al. (2012)
Biological	15	CA Pigs	3 – 9	53 – 74	-	↑	=	-	-	Girard et al. (2012)
Biological +denitrification	16	NL Poultry	>0.36 **0.71**	76 – 93 **85** ± *8*	-62 – 74 **21** ± *50*	-541 – 0 **-283** ± *187*	-4 – 32 **12** ± *15*	47 – 73 **59** ± *9*	34 – 71 **47** ± *12*	Melse & Mosquera (2014); Melse, van Hattum, et al. (2012); Melse, Hofschreuder et al. (2012)
Biological +denitrification	17	NL Poultry	>0.47 **3.0**	54 – 92 **71** ± *13*	0 – 57 **32** ± *23*	-1445– -77 **-419** ± *532*	-1 – 8 **3** ± *2*	25 – 50 **40** ± *7*	5 – 30 **25** ± *5*	Melse & Mosquera (2014); Mosquera et al. (2012); Melse, Hofschreuder et al. (2012)
Biological +denitrification	18	NL Pigs	>1.6 **3.6**	56 – 99.6 **86** ± *15*	35 – 65 **48** ± *11*	-1099 – -388 **-643** ± *288*	-16 – 3 **-3** ± *7*	63 – 77 **69** ± *6*	-	Melse, Hol, et al. (2012); Melse & Mosquera (2014); Melse, Hofschreuder et al. (2012)

Type of air scrubber	**ID**	**Country Animals**	**EBRT (s)** range **μ ± σ**	**NH_3 (%)** range **μ ± σ**	**Odour (%)** range **μ ± σ**	**N_2O (%)** range **μ ± σ**	**CH_4 (%)** range **μ ± σ**	**PM_{10} (%)** range **μ ± σ**	**$PM_{2.5}$ (%)** range **μ ± σ**	**Reference**
Biological cellulose filter	19	DK Pigs	0.3 – 0.4	-	**56** ± *17*	-	-	-	-	Feilberg et al. (2010)
Biological (untreated) Leca®	20	DK Pigs	**4.5**	**66** ± 13		-	-	-	-	Liu et al. (2014)
Biofilter	21	NL Pigs	**10**	9 – 99 **72** ± *30*						Uenk et al. (1993)
Biofilter	22	NL Poultry	**3.3**	-60 – 93 **47** ± *62*	46 – 92 **62** ± *19*	-167– -37 **-101** ± *61*	-5 – 21 **5** ± *11*	>55 – >88 **>73**	ND	Melse & Hol (2012)
Biofilter[(c)]	23	DE Pigs	-	-40 – 36	-5 – 94	-	-	-	-	Martens et al. (2001)
Biofilter Coconut and peat	24	DE Pigs	3 - 19 **6**	-26 – 83 **15**	25 – 88 **78**	-	-	-	-	Hartung & Jungbluth (2001)
Biofilter Coconut and peat	25	DE Pigs	3 - 40 **6**	-9 – 81 **36**	58 – 95 **81**	-	-	-	-	Hartung & Jungbluth (2001)
Biofilter Woodchips	26	NL Pigs	**2.6**	7 – 83 **40** ± *32*	6 – 73 **47** ± *26*	-400 – 13 **-84** ± *123*	-43 – 41 **-9** ± *50*	>77 – 94 **90** ± *5*	>66 – >96 **>77**	Melse, Hol, et al. (2014)
Biofilter Woodchips	27	FR Pigs	6 - 15	<10 – >90	-	↑	-	-	-	Dumont, Hamon, et al. (2014); Dumont, Lagadec, et al. (2014)
Biofilter inorganic gravel	28	CA Pigs	**252**	-	-	-	**43** ± *0.6*	-	-	Girard et al. (2011)

Type of air scrubber	**ID**	**Country Animals**	**EBRT (s)** range **μ ± σ**	**NH_3 (%)** range **μ ± σ**	**Odour (%)** range **μ ± σ**	**N_2O (%)** range **μ ± σ**	**CH_4 (%)** range **μ ± σ**	**PM_{10} (%)** range **μ ± σ**	**$PM_{2.5}$ (%)** range **μ ± σ**	**Reference**
Combined W-C-BF	29	NL Pigs	**7**	99 – 100 **99** ± *1*	-	-	-	90 – 94 **93** ± *1*	81 – 95 **90** ± *2*	Aarnink et al. (2007); Melse, Hofschreuder et al. (2012)
Combined C-B	30	NL Poultry	>0.16	27 – 93 **53** ± *18*	21 – 86 **54** ± *22*	-20 – 10 **-4** ± *9*	-15 – 18 **2** ± *9*	53 – 90 **71** ± *18*	-4 – 38 **22** ± *19*	Melse et al. (2011); Melse, Hofschreuder et al. (2012)
Combined C-B[(d)]	31	NL Pigs	>0.43	93 – 99 [(e)] **97** ± *2*	-43 – 85 **26** ± *45*	-11 – 21 **5** ± *12*	-28 – 22 **2** ± *17*	37 – 60 **45** ± *8*	44	Melse et al. (2011); Melse, Hofschreuder et al. (2012)
Combined C-BF Combined C-B Combined W-C-BF	32	NL Pigs	**3** **0.6** **7.5**	**70** ± *13* **83** ± *2* **100** ± *1*	-	-	-	**81** ± *3* **61** ± *3* **93** ± *1*	**62** ± *9* **47** ± *2* **90** ± *2*	Zhao et al. (2011); Melse, Hofschreuder et al. (2012)
Combined W-B-B	33	DE Pigs	>1.13	75 – 97 **86** ± *8*	-103 – 90 **49** ± *55*	-	-	69 – 77 **74** ± *3*	81 – 96 **86** ± *7*	DLG6098 (2012)
Combined B-B-B	34	DE Pigs	>1.52	90 – 92 **91** ± *1*	51 – 90 **76** ± *11*	-	-	91 – 95[(b)] **93** ± *1*	-	DLG5955 (2010)
Combined W - BF	35	DE Pigs	>1.45	90 – 96 **93** ± *3*	72 – 92 **82** ± *6*	-	-	76 – 89[(b)] **84** ± *6*	-	DLG5944 (2010)
Combined B-B-B (cellulose)	36	DK Pigs	>0.92	**97**	**80**	-	-	-	-	Riis (2012)
Combined B-B-B (cellulose)	37	DK Pigs	**1.5**	**79** ± *7*	-	-	-	-	-	Liu et al. (2012)

W = water stage (no packing material); C = chemical scrubber; B = biological scrubber (including water scrubber with packing material); BF = biofilter
CA = Canada ; DE = Germany; DK = Denmark; NL = The Netherlands
Pi/Po = Pigs and Poultry
↑ = Increase in concentration

(a) Intermittent water recirculation
(b) Measurement of total dust instead of PM_{10}
(c) 5 different types of biofilters where tested, with different types of packing material: biochips, coconut fibre/ peat mixture, chopped bark and wood, bioContact filter pellets covered with bark, crude compost
(d) Washing water of biological section (B) is reused in chemical section (C)
(e) Two measurements when acid tank was empty were not taken into account

all types of air scrubbers (Table 1.3). Part of this variation may be explained by the relatively large uncertainties associated with olfactometric measurements. In the study of Jonassen et al. (2012), 16 samples of a 3-stage combined air scrubber were analysed by three laboratories using the international standard CEN EN 13725 (2003). The odour removal efficiency differed from 16% to 80%, indicating that the result of the measurement strongly depends on the laboratory. Melse & Mol (2004) found that 80% of the variations in odour removal efficiency of a biofilter was attributed to variations in the biofilter performance, 20% was attributed to the olfactometric method.

A low odour reduction is expected in chemical air scrubbers, as the low pH-value prevents colonization of the packed bed by microorganisms (Hartung, 2008). However, Table 1.3 does not show a significantly lower odour reduction for the chemical scrubbers as compared to the other types of air scrubbers. Some bacteria like the chemolithotrophic Thiobacillus ferrooxidans and Thiobacillus thiooxidans that oxidize hydrogen sulphide, have a pH optimum below 4 (Syed, Soreanu, Falletta, & Béland, 2006) and could therefore survive in a chemical scrubber. This may explain why still a relatively high odour reduction can be found in a chemical air scrubber. Some scrubbers even produce odour instead of removing it, although the hedonic tone of odour can change (Belzile et al., 2010). The measured removal of odour by olfactometry is the sum of the removal of many different odorous components. The odour removal efficiency can decrease at constant odour load if a relative increase occurs in an odour component which is difficult to remove in comparison to other odour components (Melse & Mol, 2004). Chemical analysis of odorants (Table 1.4) at biological air scrubbers and biofilters with cellulose and Leca® (Light-expanded clay aggregates) packing material, shows that less volatile and water-soluble compounds (e.g., phenols, carboxylic acids, and indoles) are removed relatively efficiently with removal efficiencies above 75% (Feilberg et al., 2010). The removal of reduced organic sulphur compounds (e.g. dimethylsulphide) in these systems is more difficult, even increases in concentrations were noticed. The latter is linked to the formation of anaerobic zones within the packing material that could lead to the formation of reduced

sulphur compounds from other sulphur-containing organic compounds (e.g., sulphur-containing dust-borne proteins) (Feilberg et al., 2010).

Table 1.4. Reported removal efficiencies of some important odorants in air scrubbers at pig housing facilities.

Type of air scrubber	**H_2S (%)** range **mean** ± *stdev*	**Dimethyl sulphide (%)** range **mean** ± *stdev*	**Acetic acid (%)** range **mean** ± *stdev*	**p-cresol (%)** range **mean** ± *stdev*	**Indole (%)** range **mean** ± *stdev*	**Reference**
Biological cellulose filter	-	**-10** ± *34*	**87** ± *3*	**92** ± *2*	**93** ± *3*	Feilberg et al. (2010)
Combined B-B-B (cellulose)	**22** ± *8*	**-69** ± *52*	**97** ± *3*	**90** ± *10*	**89** ± *6*	Hansen, Liu, et al. (2012)
Combined B-B-B (cellulose)	30 – 90	<20	-		-	Feilberg et al. (2012)
Combined B-B-B (cellulose)	76	10	99.6	98	ND	Hansen, Liu et al., (2012); Jonassen et al. (2012)
Combined B-B-B (cellulose)	**75** ± *11*	**15** ± *16*	**99** ± *1*	**98** ± *1*	**95** ± *1*	Liu et al. (2012)

B = biological scrubber (including water scrubber with packing material)

1.5.2.3 *Nitrous oxide and methane*

The removal of nitrous oxide and methane in chemical air scrubbers is difficult as these gases have a low water solubility compared to ammonia (Henry coefficient of respectively 0.03 $mol.kg^{-1}.bar^{-1}$, 0.001 $mol.kg^{-1}.bar^{-1}$; Lide & Frederikse, 1995 and 71 $mol.kg^{-1}.bar^{-1}$ for ammonia; Dasgupta & Dong, 1986). Instead of a reduction of nitrous oxide, all the measurements at biological air scrubbers presented in Table 1.3 show an increase of this pollutant, even up to 400% [ID11]. It is known from waste water treatment that N_2O can be formed during nitrification, denitrification and by the chemical reaction of nitrite and hydroxylamine (Kampschreur, Temmink, Kleerebezem, Jetten, & van Loosdrecht, 2009). Scrubbers with a short residence time (less than 2 seconds), as monitored by Mosquera et al. (2011), showed an increase of 69.8 ± 42.3% [ID10]. Those with a longer residence time showed an average increase of 207.8 ± 154.0 % [ID11], meaning that a longer residence time can have a negative impact. Denitrification further increases the risk of N_2O formation as it is an intermediate product in the denitrification reaction (Eq.1.5). The biological air scrubbers with denitrification step, described in Table 1.3, had a N_2O production rate ranging from 17% to 66% of the incoming NH_3-N (Melse & Mosquera, 2014; [ID16, ID17 and ID18]). This is high compared to municipal wastewater treatment plants applying the nitrification-denitrification system, where this rate typically lies between 0 to 14.6% according to different studies (Daelman, van Voorthuizen, van Dongen, Volcke, & van Loosdrecht, 2013; Kampschreur et al., 2009; Yoshida, Mønster, & Scheutz, 2014). Table 1.3 shows that also biofilters have the risk of producing N_2O since nitrifying bacteria are present. Dumont, Lagadec et al. (2014) found that 10 to 40% of the incoming NH_3-N was converted to N_2O, with a maximum N_2O production of about 1 $gN_2O.m^{-3}.h^{-1}$, and attributed this to incomplete denitrification in stagnant zones depleted of oxygen. At two combined scrubbers taken up in Table 1.3 no obvious removal or production of N_2O was measured [ID30 and ID31], implying that nitrifying bacteria were not present, probably caused by the breakthrough of acid to the biological section.

The individual methane removal efficiencies in Table 1.3 at chemical and biological air scrubbers vary around zero. Some biofilters were particularly designed to remove methane using methane oxidising bacteria or methanotrophs (MOB). Most studies with these bacteria are found on biofilter using compost or soil as organic filter material rather than wood chips (Melse & Van der Werf, 2005; Nikiema et al., 2005; Pawłowska, Rożej, & Stępniewski, 2011). However, Girard et al. (2011) investigated the removal of methane for concentrations applicable in pig housing facilities with an inorganic filter material of gravel and found an average removal of 43% [ID28]. Ramirez et al. (2012) could increase this removal up to 65% by adding non-ionic surfactants. For sufficient methane removal by biological activity, an EBRT of several minutes is required (Girard et al., 2011; Melse & Van der Werf, 2005).

1.5.2.4 *Particulate matter*

On average, the removal efficiency of PM_{10} and $PM_{2.5}$ for chemical and biological air scrubbers lies around the imposed values from guidelines or legislation Table 1.2. It can be seen in Table 1.3 that the removal of PM_{10} is slightly higher than that of $PM_{2.5}$ indicating that larger particles are more easily removed. Again, a large variation in the measured values can be observed. Melse, Hofschreuder, & Ogink (2012) reported that even a net production of fine dust can be found in chemical air scrubbers with EBRTs lower than 3 seconds due to the formation of salt aerosols. This salt aerosol production is hypothesized to play a role in the total emission of fine dust. The removal of fine dust by biofilters was investigated by Melse & Hol (2012), reporting a good average removal efficiency of 73% [ID22]. Nevertheless, the remark must be made that removing high dust concentrations with only a biofilter, induces a high risk of clogging. The removal efficiency of PM_{10} and $PM_{2.5}$ is relatively high and more constant in combined air scrubbers, as more scrubber stages will result in a higher removal efficiency due to inertial impaction.

1.5.3 Monitoring removal efficiencies

A rigorous comparison of the removal efficiencies reported in Table 1.3 is not straightforward. Not only do they depend on the technical aspects of the installation, the management and maintenance of the air scrubbers, also the applied measuring technique can differ. Moreover, most data provided in Table 1.3, are obtained from point measurements, reflecting a limited time frame. Therefore, the use of continuous measuring system to monitor the concentration difference of a target component (e.g. ammonia) between the in- and outlet of air scrubbers and biofilters, in combination with an automatic data logging system, would provide a more reliable assessment of the scrubber's performance. This would not only allow for a better follow-up of the performances as part of the overall farm management system (e.g. detecting the need for maintenance), but also benefit regulatory services in view of control activities with regard to permits. Laser-based closed path optical measuring systems could be used for this purpose, as they are less sensitive to drift phenomena as compared to electrochemical instruments (Mosquera et al., 2014). Furthermore, these systems are expected to become much cheaper in the near future, especially if the demand for such equipment would increase. More research on the applicability of these systems on air scrubbers and biofilters is therefore recommended.

1.5.4 Influence of disturbance variables

Part of the variation in Table 1.3 can be explained by the main disturbance variables which are airflow rate, pollutant load and relative humidity of the incoming air and temperature. The airflow rate through the scrubber is determined by the ventilation rate in the housing facility. An increased ventilation rate will lower the gas residence time in the air scrubber, decreasing the available contact time for mass transfer (EBRT) and thus resulting in a lower removal efficiency. This effect particularly holds for less water soluble components. Melse & Ogink (2005) put forward a minimum EBRT for efficient ammonia removal of 0.4 to 1.1 s and of 0.5 to 2.3 s for odour removal for chemical and biological air scrubbers, respectively. For sufficient methane

removal by biological activity, an EBRT of several minutes is required (Girard et al., 2011; Melse & Van der Werf, 2005).

Besides the ventilation rate, also the incoming pollutant concentration affects the scrubber performance. Hadlocon, Manuzon & Zhao (2014) showed that the removal efficiency of an acid spray scrubber (without packing), logarithmically decreased with increasing ammonia concentration at the inlet. In a biological air scrubber, a constant ammonia removal efficiency can be found with increasing ammonia loading, until the maximum capacity of the biological community is reached (Xue, Wang, Wu, Zhang, & Xie, 2010). Sudden changes in the ammonia load affect pH and the level of free HNO_2, and cause an immediate (within few hours) change in ammonia consumption rate of the bacteria (Ottosen et al., 2011). However, at persisting load change below inhibitory concentrations (over a period of weeks) the biomass can adapt to the new average NH_3 load (Ottosen et al., 2011).

An increased relative humidity of the incoming air leads to a lower evaporation rate in air scrubbers (Seaward, Segall, Ott, & Donatelli, 1984), affecting the operational costs. In biofilters, an increasing relative humidity has a positive effect on the removal efficiency as it wettens the filter material (Dumont, Hamon, et al., 2014). Ottengraf (1986) recommended a bed moisture content between 40 and 60% to avoid dry out leading to gas channelling on the one hand and to prevent excessive watering, reducing the available specific surface area and causing impaction of the bed, on the other hand (Delhoménie & Heitz, 2005).

Temperature is expected to have a direct negative effect on the removal efficiency of all gaseous pollutants in air scrubbers as it influences their water solubility (Henry's law constant). The water solubility of ammonia decreases by 5% for a temperature rise of 1°C (Sander, 1999). However, statistical analysis through analysis of covariance (ANACOVA) with air temperature as the covariate variable, did not show a significant effect of air temperature on the efficiency of an acid spray scrubber ($p = 0.82$) for temperatures ranging from 12°C to 30°C (Hadlocon, Manuzon, et al., 2014). On the other hand, Melse, Ploegaert, et al. (2012) noted a daily pattern in the removal efficiency

of a biological air scrubber with varying airflow rate because of varying temperature and concluded that the decreased solubility of ammonia at higher temperatures during the day was probably the main reason of this variation. The water solubility for H_2S, CH_4 and N_2O decreases with about 2.5% for a temperature rise of 1°C (De Bruyn et al., 1995; Kavanaugh & Trussell, 1980; Lide & Frederikse, 1995).

1.6 Process design options for air scrubbers

In order to cope with the effect of disturbance variables, which depend on the indoor animal production processes, the design and operation of air scrubbers must be considered thoroughly. Important design criteria concern the air and liquid flow configuration of the scrubber, the packing dimensions and packing material.

1.6.1 Air flow configuration

The air flow configuration of an air scrubber comprises the type of fans used and the flow direction through the packing. The fans providing the flow through the scrubber need to be powerful enough to overcome the additional pressure drop created by the scrubber (Arends et al., 2008). Fans are mostly located in the housing facility, in front of the scrubber. In some cases they can be placed behind the scrubber causing higher outlet air velocities and resulting in more intense mixing and dilution of the outgoing air (Van linden et al., 2011). This may be beneficial for preventing odour nuisance to surrounding residential areas. However, this configuration will not be applied for a chemical air scrubber, because the accidental breakthrough of acid droplets would cause corrosion. To reduce the energy consumption of the fans, frequency controlled fans can be used, which are only switched on when the required ventilation rate reaches a certain level (InfoMil, 2013).

Regarding the flow direction of the exhaust air through the packing material, typically cross-current or counter-current flow configurations result in a higher removal efficiency than co-current scrubbers. The counter-current air scrubbers provide the highest theoretical removal efficiency as they maximize

the average driving force for absorption, i.e. the concentration difference between liquid and gas phase, throughout the scrubber. This is particularly important for the removal of less water-soluble components like odour, N_2O and CH_4. Cross-flow air scrubbers have a shorter residence time compared to counter-flow air scrubbers for a given set of dimensions (Albright, 2009), which has a negative impact on the removal efficiency. However, cross-flow air scrubbers also have a lower pressure drop and require a lower liquid-to-gas ratio than in counter-current design (Albright, 2009). The amount of liquid injected into the scrubber per given volume of gas flow, needed to thoroughly 'wet' the scrubber, is thus lower. A cross-flow configuration is less sensitive to uneven water distribution over the package. Even when a small area of the packing is not wetted, e.g. due to malfunctioning of a spray nozzle, the exhaust air will still flow horizontally through other wetted parts of the packing material. In contrast, when a counter-flow scrubber spray nozzle malfunctions, the resultant dry spot will tend to propagate deep into the packing bed, causing severe malfunctions (Eldridge, 2008).

1.6.2 Liquid flow configuration

Distributing the washing water equally over the packing material is very important to avoid dry spots, which could result in a bypass of untreated air leaving the scrubber. The washing water is sprinkled homogeneously at the top of the packing material with spray nozzles. The system must be resistant to plugging and fouling, providing free space for the gas flow while allowing operating flexibility (Arends et al., 2008). The smaller the droplets, the larger their specific surface area, leading to increased removal efficiencies. On the other hand, smaller droplets are more difficult to remove from the airstream by the demister installed behind the scrubber (Manuzon, Zhao, & Jonjak, 2011). In terms of energy requirements, the demister implies an extra pressure loss, which can even exceed the pressure loss caused by the packing as such (Arends et al., 2008). A daily automatic cleaning system for the demister is therefore often used, especially for larger installations (DLG5957, 2011; VDI 3478 Part 1, 2011).

1.6.3 Packing dimensions

The required volume of the packing of the air scrubber is based on the maximum ventilation rate applied in the animal housing facility and the appropriate minimum EBRT. The true (actual) residence time, which takes into account the exact porosity of the packing material, is mostly not known (Waweru, Herrygers, Van Langenhove, & Verstraete, 2008). A smaller true residence time than the EBRT is found when dirt accumulates in the pores. However, adsorption of the pollutants to the packing material can increase the true residence time. In Flanders, the minimum EBRT is not fixed by legislation but set by suppliers based on experience. The removal of less soluble components like odorants and greenhouse gases requires longer EBRTs and thus larger air scrubber installations for the given incoming gas flow rate.

The air velocity through the scrubber is determined by the gas flow rate and cross section area. The VDI guideline (VDI 3478 Part 1, 2011) recommends velocities between 0.3 and 2.5 $m.s^{-1}$. The dimensions of the air scrubber packing also influence the pressure drop, which is proportional to the packing height and the square of the air velocity (Melse, van Wagenberg, & Mosquera, 2006). By increasing the cross section area of an air scrubber for the same volume, a lower air velocity is achieved, reducing the travelling distance through the packing for the same contact time and lowering the pressure drop.

1.6.4 Packing material

The choice of the packing material is very important. A high specific surface area, typically between 100 and 500 $m^2.m^{-3}$ of clean and dry packing (Arends et al., 2008) is required to obtain a high removal efficiency. However, an increasing surface area is typically accompanied by a decreasing pore size, leading to a higher pressure drop (Delhoménie & Heitz, 2005). Once in operation, the pressure drop is further increased by accumulation of dust, bacterial growth and salt deposition on the packing material, resulting in higher energy requirements. The specific surface area is thereby lowered as well. A pressure drop exceeding 400 to 500 Pa per m of packing material at a gas velocity of 0.1 $m.s^{-1}$ indicates clogging (VDI 3478 Part 2, 2008). In practice, a

relatively small increase in pressure drop can already indicate operational problems. It is therefore required to clean the packing material at least once a year and to measure the pressure drop, e.g. with an orifice (InfoMil, 2013). Completely leaving out the packing material results in a minimal pressure drop, reducing the electricity cost. Such test was applied on a prototype acid spray scrubber during one year (Hadlocon, Zhao, Manuzon, & Elbatawi, 2014; Manuzon, Zhao, Keener, & Darr, 2007), still reaching an average ammonia removal efficiency of 88% (Hadlocon, Zhao, et al., 2014). This is a promising process option for chemical air scrubbers at animal housing facilities, in order to avoid clogging of the packing. However, it does not prevent clogging of the spray nozzles (due to high dust loadings in the incoming exhaust air), which could significantly reduce the contact area between the droplets and the air and in this way negatively affect the removal efficiency of the scrubber. Results in full scale applications without packing material are necessary to fully appraise these types of installations.

The packed filter of an air scrubber is mostly made of plastic, which is less susceptible to collapsing and clogging than organic biofilter material. More recently, impregnated cellulose has been applied at biological air scrubbers (Feilberg et al., 2010), as it remains wet for a longer time and has the advantage to fix the biofilm more strongly. However, this material may have a higher risk of collapsing. Other types of packing material are applied to remove specific components. For instance, recent experiments with Leca® showed that high removal of both H_2S and methanethiol from pig exhaust air could be obtained if the air velocity was kept sufficiently low (Feilberg, Liu, Pedersen, & Hansen, 2012; D. Liu, Løkke, Riis, Mortensen, & Feilberg, 2014). This is assumed to be achieved by iron oxide, found at the active surface, which is involved in the non-biological degradation of sulphur compounds (Gendel, Levi, & Lahav, 2009; Terörde, van den Brink, Visser, van Dillen, & Geus, 1993). However, attention should be paid concerning the formation of N_2O when Fe is present. Kampschreur et al. (2011) found that nitric oxide and nitrous oxide were formed during chemical nitrite reduction by ferrous iron (Fe(II)). This pathway can be a significant cause of N_2O formation, especially in systems where nitrite

is present at elevated concentrations (Kampschreur et al., 2011). The reduction of methane has been reported on biofilter material like compost and gravel as this is the preferred habitat of methane oxidising bacteria (MOB) (Hanson & Hanson, 1996). In nature, MOB present in soil and water bodies eliminate 95% of the total emitted methane in the environment (Ramirez et al., 2012). The conditions for growth of these bacteria on other types of packing material needs to be further investigated.

1.7 Process control options

For reliable operation of air scrubbers, the water flow rate or trickling density, electrical conductivity (EC) and pH in the washing water need to be controlled. The dosage of solvents and the inoculation of bacteria provide perspectives to increase the removal efficiency of the less water soluble components (odorous compounds, methane, nitrous oxide).

1.7.1 Water flow rate - trickling density

One of the main influencing operating parameters for chemical and biological air scrubbers is the trickling density, which is the flow rate of washing water sprayed over a unit of specific packing area. A suitable trickling density is required to reach the appropriate liquid-to-gas ratio. For dust removal, the required liquid-to-gas ratio is a function of the mechanical design of the air scrubber. Regarding gas absorption, a higher liquid-to-gas (L/G) ratio is required to remove less soluble gases. The maximum (L/G) in countercurrent flow is limited by flooding, the minimum is fixed by the minimal amount to wet the entire packing with a thin liquid film (US EPA, 1990). In practice, (L/G) ratios are usually set at 1.1 to 1.7 times this minimum rate (Schnelle & Brown, 2002b). The estimated (L/G) of the acid spray scrubber was 2.10^{-4} at an air residence time of 0.43 seconds (Hadlocon, Zhao, et al., 2014). Chemical and biological air scrubbers require a trickling density between 1.7 and 2.2 $m^3.h^{-1}$ per m^2 cross sectional area according to Arends et al. (2008). According to the VDI guideline for biological air scrubbers, the trickling density must be less than 5 $m^3.h^{-1}.m^{-2}$ (VDI 3478 Part 2, 2008) to prevent that the biofilm is washed off by shear stress. Most biological air scrubbers are even designed at

a much lower trickling density of 0.8 $m^3.h^{-1}.m^{-2}$. Chemical air scrubbers typically operate at a higher trickling density of around 10 $m^3.h^{-1}.m^{-2}$. The trickling density in a bioscrubber ranges between 10 and 60 $m^3.h^{-1}.m^{-2}$ (VDI 3478 Part 1, 2011).

Higher inlet concentrations can be dealt with by increasing the trickling density, both in chemical and biological air scrubbers (Arends et al., 2008; Hadlocon, Manuzon, et al., 2014; Xue, Wang, Wu, Sun, & Xie, 2010). However, if the liquid film on the surface of the biofilm becomes too thick, the mass transfer resistance will increase, eventually decreasing the elimination capacity for low water soluble components (Xue, Wang, Wu, Sun, et al., 2010). An optimum trickling density must thus be found. It should as well be noted that an increase in the trickling density entails an increase in the gas flow resistance, thus an increase in pressure drop (VDI 3478 Part 1, 2011). In practice, the water flow rate is typically kept constant despite variations in the incoming gas flow rate and pollutant load. Adjusting the water flow rate to the incoming load at high loading rates, opens perspectives for keeping a high removal efficiency at all times. Estrada et al. (2014) enhanced the mass transfer in a standard biotrickling filter for methane removal by applying high liquid recycle rates and an innovative gas recycling strategy, increasing the turbulence in the gas phase while keeping an overall or nominal EBRT of 240 s (effective EBRT of 12.6 s), reaching an elimination capacity as high as 30 $gCH_4.m^{-3}.h^{-1}$. This illustrates how innovative control options can increase the removal rate of a less water-soluble component.

1.7.2 Water discharge - EC control

The decisive factor for proper operation of air scrubbers is the removal by discharge of components accumulated in the washing water (Arends et al., 2008). This is typically done based on the nitrogen concentration in the water. Since direct nitrogen measurements are too expensive, typically electrical conductivity (EC) or density is measured instead (InfoMil, 2013). In a biological air scrubber, the EC measurement was shown to correlate well with the sum of present ammonia, nitrite and nitrate in $gN.L^{-1}$ (Melse, Ploegaert, et al., 2012). Based on empirical observations and theoretical calculations, the

combined concentration of NH_4^+ and NO_x^- per $mS.cm^{-1}$ of conductivity was found to be approximately 7 mM (Ottosen et al., 2011). Keeping the total nitrogen concentration below 3.2 $gN.L^{-1}$, this implies that the EC value must be kept lower than 16 $mS.cm^{-1}$. However, preventing inhibition by FA and FNA is not so straightforward, many different inhibition thresholds being reported in literature (Anthonisen, Loehr, Prakasam, & Srinath, 1976; Buday, Drtil, Hutňan, & Derco, 1999; Carrera, Jubany, Carvallo, Chamy, & Lafuente, 2004; Tan et al., 2008). Ottosen et al. (2011) suggested a maximum EC value of 20 $mS.cm^{-1}$. The EC value of a chemical air scrubber is mostly kept below 250 $mS.cm^{-1}$ to stay below the maximum solubility of ammonium sulphate (InfoMil, 2013). Conductivity measurements are not always stable, because of the influence of evaporation and the disturbance of the conductivity of the dosed acid. A density measurement is indicated by InfoMil (2013) as an alternative way of monitoring the accumulation of nitrogen components.

Even though important with respect to inhibition of bacteria (Anthonisen et al., 1976) and formation of N_2O (Kampschreur et al., 2009), nitrite as such is typically not monitored and taken into account for discharge control. Due to the high ammonia loads, the nitrite concentration in air scrubbers is rather high compared to WWTP leading to inhibition of NOB and thus incomplete nitrification (Melse & Ploegaert, 2011; Ottosen et al., 2011; Wett & Rauch, 2003). Besides, the interaction between different components needs to be taken into account as well. For instance, high ammonium and nitrite concentrations in the washing water lead to decreased methane removal efficiencies (King & Schnell, 1994; Veillette, Viens, Ramirez, Brzezinski, & Heitz, 2011). So if one aims for removing both ammonia and methane, ammonia should be removed in a first stage, only introducing MOB in a second stage where nitrogen concentrations are relatively low.

1.7.3 Acid dosage - pH control

Sulphuric acid (H_2SO_4) is typically used in chemical air scrubbers to lower the pH as it is relatively cheap. The formed ammonium sulphate ($(NH_4)_2SO_4$; Eq.1.2) can be used as a fertilizer. However, this application can be hampered by the varying composition of the discharge water and the need for dedicated

land application equipment (Meers et al., 2013). The use of other acids than sulphuric acid for the removal of ammonia was investigated by Starmans & Melse (2011). Citric acid seems to be promising as this organic acid is safer to handle than sulphuric acid, however it has the disadvantage to be more expensive and it has a strong odour itself. A small scale scrubber working on citric acid is already installed at a fattening pig farm in the Netherlands (Boerderij, 2013) but long term results are not yet described. To overcome problems with applying ammonium sulphate discharge water on sulphate rich soils, nitric acid could be used instead as a washing medium. Furthermore, the resulting ammonium nitrate discharge water is a high-nitrogen fertilizer. It also has a higher water solubility meaning that the amount of discharge water could be lowered. However, it is more expensive and more caution must be paid to safety as the dry form of the formed salt has its application in explosives.

In biological air scrubbers, the optimum working pH is 7.5 (Ramírez, Gómez, Aroca, & Cantero, 2009). Acidification due to a persistent low ammonia load can be prevented by keeping a sufficiently high discharge rate. The pH affects the process performance also in other ways than through the ammonia removal efficiency. A study at a biofilter operating at different pH values showed that the N_2O emissions significantly increased with decreasing pH (L. Yang, Wang, & Funk, 2014). This was in line with comparable studies in soil science (Firestone, Firestone, & Tiedje, 1980; B. Liu, Mørkved, Frostegård, & Bakken, 2010). However, it conflicts with findings at a WWTP where the highest N_2O emissions were found at a pH of 8.5 and the lowest at a pH of 6 (Kampschreur et al., 2009). Further research is necessary as these processes are not yet completely understood.

1.7.4 Dosage of solvents

The removal of poorly water soluble components in a biological air scrubber can be improved by addition of an organic solvent, increasing the mass transfer and thus its availability for bacteria (Cesário, 1997; Volckaert, 2014). Lebrero et al. (2013) demonstrated very high removal efficiencies (>96%) of four types of VOCs with different hydrophobicity present in trace level concentrations, in a two-phase biotrickling filter in which 30% of the trickling medium was

taken up by silicone oil. This technique can be interesting to increase the removal efficiency of methane. Ramirez et al. (2012) could increase the removal efficiency of methane in a biofilter from 35% to 65% by adding non-ionic surfactants. The latter decrease the surface tension of a liquid, by altering the molecular forces at the liquid–gas or liquid–liquid interfaces. However, adding solvents will increase the operational costs, both concerning the solvent as such and the discharge of the washing water.

The dosage of specific solvents can also be used in the reduction of possible spreads of diseases through housing emissions. The outbreak of highly infectious viral diseases like swine fever or avian influenza can be prevented by adding peracetic acid to the washing water in a chemical air scrubber (Aarnink, Landman, Melse, & Huynh, 2005). Peracetic acid was capable of reducing the bacteria and virus emissions below detectable levels. However, since its dosage is very expensive, this is an option only in times of high risk of infecting other farms.

1.7.5 Inoculation with bacteria

Even though inoculation, typically with activated sludge, is not really necessary to establish the growth of nitrifying microorganisms in a biological air scrubber, it can decrease its start-up period (Xue, Wang, Wu, Zhang, et al., 2010). Besides, the inoculation of the air scrubber with specific bacteria for the reduction of odorants and greenhouse gases is found useful. Some studies can be found where specific heterotrophic bacteria where inoculated to promote odour reduction (Jiang, Yan, & Tay, 2009; López et al., 2013; Smet, Van Langenhove, & De Bo, 1999). Juhler et al. (2009) demonstrated that the heterotrophic bacteria, which reduce odour, and nitrifying bacteria compete with each other for space and oxygen. Due to the significantly lower specific growth rate and lower dependency on oxygen, nitrifiers can only establish persistent populations where the heterotrophs are limited by substrate and not by oxygen. In cases of high organic loading, oxygen depletes within the highly active heterotrophic surface layer, thus outcompeting AOB from the biofilm. Both Kristiansen et al. (2011) and Juhler et al. (2009) emphasize that the influence of filter design and operating conditions on the key microorganisms

should be further studied to improve the odour removal efficiency and stability of these systems. Biological air scrubbers treating methane are typically inoculated with methane oxidising bacteria (Estrada et al., 2014; Girard et al., 2011). However, the low methane concentration in the animal house exhaust air may lead to substrate limitation, which negatively affects the methane removing capacity of an air scrubber, also after inoculation, as demonstrated by Ganendra et al. (2013).

The addition of nutrients or trace elements to the washing liquid can be useful to promote the growth of bacteria. While not necessary for ammonia removal (Prado, Redondo, Lafuente, & Gabriel, 2009), the growth of methane oxidising bacteria can be successfully promoted by adding Cu^{2+} (by $CuSO_4$) in high concentrations to the washing water (Estrada et al., 2014).

1.8 Conclusions and perspectives

Currently installed chemical, biological or combined air scrubbers do not always reach the required removal efficiencies for ammonia, odour and particulate matter. This was made clear in this review by confronting the legally required removal efficiencies with those realized in practice and agrees with the findings of Lemay et al. (2009), stating that an air cleaning technology for effective reduction of all main air contaminants coming from pig buildings could not yet be found. However, current air scrubbers should perform properly for ammonia reduction. More control of the full-scale installations is recommendable to ensure proper operation and maintenance of these systems. The use of closed path laser-based optical measuring devices as part of a continuous monitoring system on air scrubbers and biofilters could be beneficial in this respect. The reduction of greenhouse gases such as nitrous oxide and methane is not yet mandatory, but it seems recommendable to be pro-active. The formation of nitrous oxide in biological air scrubbersis likely and warrants further research to elucidate the underlying mechanisms. It is advised to take up measurements of nitrous oxide as part of monitoring campaigns on biological air scrubbers to get more insight, and in view of the development of mitigation strategies. Process options for methane removal in

air scrubbers exist but require further optimization to reduce the pressure of animal housing facilities on the environment.

Until now, the design of air scrubbers at pig and poultry housing facilities was mostly based on the removal of ammonia. Optimizing design and operation of air scrubbers should facilitate the simultaneous reduction of odour, nitrous oxide, methane and particulate matter in an efficient and cost-effective manner. With respect to design, the air and liquid flow configuration, packing dimensions and packing material should be carefully considered. Efficient removal of less soluble components like odorants and greenhouse gases, requires longer EBRTs, implying larger installations. New types of packing material, such as Leca® containing iron oxide, or even gravel to promote the growth of methane oxidizing bacteria, are promising for the removal of odour and methane but further research is needed to allow implementation in animal housing facilities.

Control options for water flow rate, water discharge and acid dosage could be optimized. An adjustable trickling density would be an interesting option to keep a sufficient high liquid-to-gas ratio at high ventilation rates. Discharge of the washing water is now controlled by electrical conductivity or density measurements which serve as an indication for the total nitrogen content. For biological air scrubbers, this leads to large discharge volumes, which could be avoided by finding a better way to measure and/or to avoid the accumulation of inhibition components. The largest bottleneck for the removal of odour or methane is the low solubility of these components. Better removal efficiencies could be obtained by the addition of apolar solvents to increase the mass transfer of these components.

2

Objectives and outline

2.1 Overall objective

This doctoral research work deals with the design and operation of chemical, biological and combined air scrubbers for the removal of ammonia from pig housing facilities. Besides ammonia, attention is paid to emissions of the strong greenhouse gases nitrous oxide and methane and to emissions of hydrogen sulphide as an important odour component. The optimization of air scrubbers is investigated combining modelling and simulation with full-scale measurements.

The main objectives of this doctoral research project are:

- The set-up of mechanistic models for chemical and biological air scrubbers, which are not limited in use for the agricultural sector but can be widely applied. Temperature and relative humidity are taken into account, as these two are interrelated with the removal efficiency.
- To investigate the influence of disturbance variables (e.g. temperature, relative humidity, ventilation rate), process design (e.g. packing dimensions, air and liquid flow configuration) and control variables (e.g. pH, discharge rate, recirculation rate) on the performance of air scrubbers.
- To gain insight in the start-up of biological air scrubbers and the influence of inoculation of biological air scrubbers as cheap and easy method to optimize the operation of these type of air scrubbers.

2.2 Outline of the thesis

In **Chapter 3,** three short-term measurement campaigns conducted at a chemical, biological and two-stage biological air scrubber installed at pig housing facilities in Flanders are addressed to investigate diurnal fluctuations in the removal performance of air scrubbers. The data of these monitoring campaign serve for the model calibration in **Chapter 4**.

Mechanistic models for a chemical and biological air scrubber are presented in **Chapter 4** and **Chapter 5,** respectively. In the chemical air scrubber model in

Chapter 4, special attention is paid to the correlations of the mass transfer coefficient, as this is one of the most important parameters in absorption models. In the mechanistic model of the biological air scrubber in **Chapter 5,** biomass and biological conversion reactions are introduced. The pH is both assumed constant and dynamically modelled. Each model is validated using experimental data and a simulation study is performed to investigate the influence of typical prevailing disturbance variables, design parameters and operational variables.

In **Chapter 6,** the results of a long-term measurement campaign at two full scale biological air scrubbers will be described; in which the influence of inoculation on the performance of the air scrubbers in terms of ammonia removal and nitrous oxide production will be investigated. The microbial community structure of the biofilm and washing water during the measurement campaign is addressed in **Chapter 7** to get more insight in which bacteria are present.

Chapter 8 offers some final considerations, conclusions and perspectives about the design and operation of chemical, biological and combined air scrubbers.

3

Measurement campaign at three different types of air scrubbers

3.1 Abstract

This contribution presents the results from measurement campaigns conducted at a chemical, a biological and a two-stage biological air scrubber installed at pig housing facilities in Flanders. Ammonia, nitrous oxide and methane at the inlet and outlet of the air scrubbers were monitored on-line during one week using a photoacoustic gas monitor, which allowed to investigate diurnal fluctuations in the removal performance of air scrubbers. Additionally, the homogeneity of the air scrubbers, normally checked by gas detection tubes, was investigated in more detail using the continuous data. The biological air scrubber with extra nitrification tank performed well in terms of ammonia removal (86 ± 6%), while the two-stage air scrubber suffered from nitrifying bacteria inhibition. In the chemical air scrubber, the pH was not kept constant, lowering the ammonia removal efficiency. A lower ammonia removal efficiency was found during the day, when the ventilation rate was the highest. Nitrous oxide was produced inside the biological and two-stage scrubber, resulting in an increased outlet concentration of more than 200%. Methane could not be removed in the different air scrubbers because of its low water solubility.

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3.2 Introduction

Measurements at animal housing facilities (Mosquera et al., 2011) show that the ammonia removal efficiency of 70%, which is legally required in Flanders (MB31/05/2011), is not always reached in practise. Furthermore, there is a growing international concern for the livestock related greenhouse gases methane and nitrous oxide (Gerber et al., 2013). Still little attention is being paid to the performance of air scrubbers in terms of the reduction or production of these greenhouse gases. Measurements with high time resolution are necessary to follow the dynamics and to relate the emission pattern with underlying processes affecting the emission (Mosquera et al., 2014). However, a lot of studies present only single measurements of concentrations and removal efficiencies averaged over one hour, mostly only focusing on ammonia (Girard, Belzile, Lemay, & Feddes, 2012; Melse & Mol, 2004; Melse & Ogink, 2005; Mosquera et al., 2011; Zhao et al., 2011). Melse, Ploegaert, et al. (2012) already recognized that continuous measurements are needed to investigate day-night variations and fluctuations in the removal performance and performed a study at a biological air scrubber. Furthermore, to our knowledge, no continuous measurements of nitrous oxide and methane at air scrubbers were performed until present.

The aim of this study is to investigate the fluctuations in removal efficiency and concentration for ammonia, nitrous oxide and methane of a chemical, a biological and a two-stage air scrubber placed at commercial pig houses. The incoming and outgoing concentrations of ammonia, nitrous oxide and methane were continuously measured during one week to get insight in the performances of the respective air scrubbers.

3.3 Materials and methods

3.3.1 Description of the air scrubbers

The study was conducted on three air scrubbers placed at different pig housing facilities, located in Flanders, Belgium. The main design characteristics can be found in Table 3.1. The scrubber packings were made from inert plastic.

The chemical air scrubber (Figure 3.1) is installed at a commercial fattening pig barn with 1344 animal places (Asse, Belgium). The pH of the washing water was controlled below 4, until the maximum electrical conductivity setpoint of 180 $mS.cm^{-1}$ is reached, to stay below the maximum allowed ammonium sulphate concentration in the washing water of 2.1 $mol.L^{-1}$ or 58.6 $gN.L^{-1}$ (MB31/05/2011) set in Flanders. At that point, sulphuric acid is no longer dosed until the pH reaches 7 and the washing water is discharged to a separate storage tank. This is to obtain a neutral ammonium sulfate discharge product which can be used for field applications.

Figure 3.1. Outgoing gas measurements at the chemical air scrubber

The biological air scrubber (Figure 3.2) was located in Linter (Belgium), treating the exhaust air of a barn housing 144 sows and 3120 piglets. The installation was extended with an additional nitrification tank with bacteria grown in granular sludge to treat the washing water before reuse. In Flanders, the maximum allowed concentration of nitrogen in the biological washing water is set to 3.2 $gN.L^{-1}$ (MB31/05/2011), which corresponds to an EC setpoint of approximately 18 $mS.cm^{-1}$. When this value is reached in the biological scrubbing section, part of the washing water is discharged towards the manure pit of the housing facility and fresh water is added.

Figure 3.2. Outgoing gas measurements at the biological air scrubber

The two-stage air scrubber (Figure 3.3), installed in Mollem (Belgium) consisted of a water stage to remove dust, followed by a larger biological stage of 1.80 m packing material with a lower specific surface area. When an EC setpoint of 15 $mS.cm^{-1}$ in the biological scrubber section (stage 2) is reached, the washing water from the water scrubbing section (stage 1) is discharged to the manure pit of the animal housing facility, the washing water from stage 2 flows to stage 1 and fresh water is added to stage 2. In this way, the washing water of section 1 can have a higher nitrogen content without inhibiting the nitrifying bacteria in stage 2. This scrubber was installed at a fattening pig housing facility of 1515 animal places.

Figure 3.3. Outgoing gas measurements at the two-stage air scrubber

Table 3.1. Design characteristics of the measured air scrubbers

Type	Maximum ventilation rate(1) ($m^3.h^{-1}$)	Ventilation rate(2) ($m^3.h^{-1}$)	Flow configuration	Packing specific surface area ($m^2.m^{-3}$)	Packing dimensions(3) (m x m x m)	Minimum EBRT(4) (s)
Chemical	80.640	60.000	countercurrent	80	7.0 x 0.3 x 2.8	0.35
Biological(5)	97.180	100.000	countercurrent	100	10.0 x 3.0 x 0.9	1.10
Two-stage(6)	121.200	120.000	crosscurrent	stage 1: 300 stage 2: 150	6.0 x 4.0 x 3.0 stage 1: 0.15 stage 2: 1.80	2.20 stage 1: 0.08 stage 2: 0.97

(1) Based on the number of animals present in the housing facility
(2) Used for air scrubber design
(3) Total length x width x height; Length of the two-stage scrubber packing is given separately as well for each packing stage, with empty space between the stages.
(4) Empty bed residence time (EBRT) of the two packings of the two-stage scrubber is given separately as well.
(5) BWL 2008.05.V1 (InfoMil, 2008a)
(6) BWL 2008.12.V1 (InfoMil, 2008b)

3.3.2 Measuring equipment

Gas concentration measurements of NH_3, CH_4 and N_2O were performed using an Innova photoacoustic gas monitor 1314 (Table 3.2) connected to a CBISS multipoint sampler (LumaSense Technologies, Denmark). The analyzer was calibrated by the manufacturer and validated with 30 ppm ammonia from a gas bottle before the measurements started. This equipment is listed as one of the prescribed measuring techniques in the VERA test protocol for air cleaning technologies (VERA, 2010), which provides a standardized protocol in Europe to test the efficiency of air scrubbers. Using a photo-acoustic monitor not only allows to measure ammonia at a high time resolution but also allows the measurement of nitrous oxide and methane at the same time and is therefore chosen in this study. However, caution must be paid when measuring low concentrations (< 2 ppm NH_3) as they could be overestimated when using this system (Mosquera et al., 2014; Osada, Rom, & Dahl, 1998). Measuring multiple gases with an infrared photoacoustic monitor can also have an effect of non-compensated interferences which could lead to under- or overestimations (Hassouna, Robin, Charpiot, Edouard, & Méda, 2013; Zhao, Pan, Rutherford, & Mitloehner, 2012). This is especially important when concentrations are measured to determine emission factors (absolute values) but is of minor importance when the removal efficiency is calculated as the difference between incoming and outgoing concentrations is used (relative values). After calibration, cross-interferences are normally compensated, allowing proper operation of the photoacoustic gas monitor.

The gas flow rate through the air scrubber, or thus the ventilation rate of the housing facility, was estimated based on continuous pressure difference measurements over the ventilation fans with a P26 differential pressure transducer (halstrup-walcher, Germany) which were linked at certain points with the estimated ventilation rate given on the climate control computer inside the housing facility. Ventilation rates are expressed as a percentage of the maximum ventilation rate. Temperature and relative humidity of the incoming and outgoing air were measured using thermocouples (EE071 probe, E+E Elektronik, Germany). The washing water of each scrubber was sampled at

least three times during each measuring campaign and analyzed in the lab for pH and EC according to EN 13037 and EN 13038. For the chemical air scrubber, the washing water was further analysed for NH_4^+ and SO_4^{2-}, for the biological and two-stage air scrubber for NH_4^+, NO_2^- and NO_3^-, using extraction and ion chromatography according to EN 13652.

Table 3.2. Specifications of the INNOVA 1314

Component	Optical filter	Detection limit [ppm]	Minimal measurement limit(1) [ppm]	Maximal measurement limit(2) [ppm]
NH_3	UA 0976	0.2	1	2 000
N_2O	UA 0985	0.03	0.15	300
CO_2	UA 0983	5.1	25.5	510 000
CH_4	UA 0969	0.4	2	4 000
H_2O	SB 0527			

(1) Set by the manufacturer as 5 times the detection limit
(2) Set by the manufacturer as 10 000 times the detection limit for 1-point compensation and 100 000 times the detection limit for 2-point compensation (only for CO_2)

3.3.3 Measuring strategy

Continuous measurements were performed on each air scrubber during one week in June 2014. Gas samples were taken sequentially at five sampling points; one before and four behind the respective air scrubber. Each sampling consisted of at least 6 consecutive measurements with a 3 minutes time interval. Only the last measurement was taken into account to overcome a possible measuring delay of the gas monitor. The sampling point before the scrubber in the pressure chamber included a dust filter to prevent dust to enter the measurement device. The sampling tubes behind the scrubber were heated to 110°C to prevent condensation. Dilution by wind was avoided by using protective ducts around the sampling points. It was chosen to sample a number of four points behind the scrubbers to compromise minimal time resolution and maximal spatial resolution. The removal efficiency was calculated using the average outgoing concentration and the subsequently measured incoming concentration. This implies a time difference between the measured incoming

and outgoing concentration of max. 1 hour and 24 min. The four measurement points were equally divided over the emitting surface area to check the variation of the outgoing concentrations in space. Air scrubbers are considered to have a spatially homogeneous removal efficiency when the relative standard deviation (RSD) of the measured outgoing ammonia concentrations over the different sampling points is lower than 30% (Mosquera et al., 2014; VITO, 2012). It gives an indication that the air velocity through the air scrubber is not homogeneous. For nonhomogeneous air scrubbers, at least 10% of the total surface must be sampled. At the beginning of each measuring campaign, homogeneity of the air scrubber was checked indicatively by measuring the outgoing ammonia concentration at least at 6 random places over the emitting surface area according to EN 15259:2007 to evaluate whether or not it is useful to start the more elaborate (and expensive) measurements with the photoacoustic monitor. This is the first step in the VERA test protocol and compendium method in Flanders to measure the efficiency of air scrubbers (Mosquera et al., 2014; VITO, 2012). Either gas detection tubes (2GTC103L ammonia low range 1-30 ppm, GASTEC Corporation, Japan) or an electrochemical ammonia sensor (MX6 iBRID, Industrial Scientific, France) were used on the biological and two-stage air scrubber. For the chemical air scrubber, this was not possible as the emitting surface area was not easily reachable. Because the accuracy of gas detection tubes and electrochemical sensors is low, the homogeneity was checked again by calculating the relative standard deviation of the four outgoing ammonia concentrations for each instance of time using the photoacoustic gas monitoring data. It was also compared with the average standard deviation over the different measurement points.

3.3.4 Calculation of additional parameters

The removal efficiency RE [%] for each pollutant is based on the incoming concentration C_{in} [ppm] and outgoing concentration C_{out} [ppm]. It was not necessary to recalculate the incoming and outgoing concentration from ppm to $gN.m^{-3}$ as the influence of the temperature difference between both on the calculation of the removal efficiency was negligible. It is calculated as follows:

$$RE = \frac{C_{in} - C_{out}}{C_{in}} \cdot 100\% \qquad \textbf{Eq.3.1}$$

It was not necessary to include the ventilation rate in Eq.3.1., despite the difference in time between the measurement at inlet air and outlet air. As the ventilation rate gradually changes, the change within this time frame is considered small enough. Additionally, the measurement of the ventilation rate has a larger uncertainty than that of the concentration and it was therefore chosen to only use the concentration.

The relative standard deviation RSD [%] was calculated for each time instance as the standard deviation SD [ppm] divided by the average $\bar{x}$ [ppm] over the 4 measurement points:

$$RSD = \frac{SD}{\bar{x}} \cdot 100\% = \frac{\sqrt{\frac{\sum (x - \bar{x})^2}{3}}}{\frac{\sum x}{4}} \qquad \textbf{Eq.3.2}$$

The time difference between the first and last measurement point was around half an hour.

The loading rate LR and removal rate RR were calculated from the incoming concentration C_{in} (ppm converted to $mg.m^{-3}$) and the removed concentration (C_{in} - C_{out}, ppm converted to $mg.m^{-3}$), respectively, multiplied by the total ventilation rate Q ($m^3.h^{-1}$) and divided by the volume of the scrubber packing $V_{packing}$ (m^3).

$$LR = C_{in} \cdot \frac{M}{\frac{R \cdot T}{p} \cdot 10^6} \cdot \frac{Q_{vent}}{V_{packing}} \qquad \textbf{Eq.3.3}$$

$$RR = \left(C_{in} - C_{out}\right) \cdot \frac{M}{\frac{R \cdot T}{p} \cdot 10^6} \cdot \frac{Q_{vent}}{V_{packing}} \qquad \textbf{Eq.3.4}$$

3.4 Results

3.4.1 Gas concentrations and removal efficiencies

Figure 3.4, Figure 3.5 and Figure 3.6 display the continuously measured data. Table 3.3 summarizes the average incoming and outgoing concentrations of ammonia, nitrous oxide and methane, their respective removal efficiency and

loading and removal rates for each type of air scrubber.

The ammonia removal efficiency in the chemical air scrubber was on average 45 ± 30% and varied between -23 ± 12% and 70 ± 9%. This means that there were periods with a negative removal efficiency or thus stripping of ammonia (Figure 3.4A). On June 17th, when the pH of the washing water was 4.02, the ammonia concentration at the chemical air scrubber decreased from 21.8 ± 1.8 ppm in the incoming air to 8.7 ± 3.1 ppm in the outgoing air, and reached a higher ammonia removal efficiency of 59 ± 15%. In the biological air scrubber, the required ammonia removal efficiency of 70% was reached throughout the measurements (85 ± 6%). The incoming ammonia concentration was rather low with values between 5.7 and 14.2 ppm (Figure 3.5A). The average outgoing ammonia concentration measured varied between 0.5 ± 0.2 ppm and 2.7 ± 0.4 ppm. In the biological air scrubber with water curtain (two-stage), the incoming ammonia concentration varied between 14.6 ppm measured at night and 23.4 ppm during the day (Figure 3.6A). The outgoing concentration measured on average 7.2 ± 0.7 ppm, with values between minimal 5.8 ± 0.9 and maximal 9.0 ± 0.9 ppm. An ammonia removal efficiency of between 52 ± 2% and 73 ± 2% was noted.

The incoming concentrations of nitrous oxide measured on average 0.11, 0.16 and 0.17 ppm for the chemical, biological and two-stage air scrubber, respectively. In the chemical air scrubber, the nitrous oxide concentration did not show an increase or decrease while flowing through the scrubber (Figure 3.4B). A higher concentration and variation between the different measuring points in the outgoing concentration could be observed at a higher ventilation rate. The outgoing nitrous oxide concentration measured on average 0.10 ± 0.2 ppm between 23:00 and 02:00 and 0.19 ± 0.3 ppm between 12:00 and 15:00. The removal efficiency varies quite significantly as well with values between -128 ± 6% and 25 ± 40%. In the biological air scrubber, a doubling in the nitrous oxide concentration could be noted going from 0.16 ± 0.06 ppm to an outgoing concentration of 0.34 ± 0.09 ppm, resulting in a production of 121 ± 64%. Four outliers in the incoming nitrous oxide concentration could be noted, with values as high as the outgoing concentration (Figure 3.5B). An even larger significant production of nitrous oxide was observed in the two-stage air

scrubber, which also includes a biological treatment stage. The incoming concentration increased from 0.17 ± 0.04 ppm to 0.49 ± 0.06 ppm, an increase with factor 3. The nitrous oxide removal efficiency varies between -307 ± 12 and 3 ± 4% (Figure 3.6B).

Figure 3.4C, Figure 3.5C and Figure 3.6C show that methane could not be removed in the chemical, biological and two-stage air scrubber with removal efficiencies of -4 ± 14%, 4 ± 9% and 5 ± 13%, respectively. A daily trend in the incoming methane concentration was observed with a higher methane concentration during the night and a lower methane concentration during the day.

Table 3.3 summarizes the calculation of the relative standard deviation for the three types of air scrubbers over the measurement period of 4 days for each air scrubber. The relative standard deviation for ammonia as for nitrous oxide is on average below 30%, but also maximum values until 57% were found. The relative standard deviation of methane was the lowest, never having a value above 25%. A trend in the relative standard deviation for ammonia, nitrous oxide and methane in time could not be found.

Table 3.3. Overview of the incoming and outgoing concentrations (C_{in} and C_{out}) of ammonia, nitrous oxide and methane and their respective removal efficiency (RE), average relative standard deviation (RSD), loading rate (LR) and removal rate (RR) for the three scrubber systems.

	C_{in} [ppm]	C_{out} [ppm]	RE [%]	RSD [%]	LR [$g.h^{-1}.m^{-3}$]	RR [$g.h^{-1}.m^{-3}$]
Chemical air scrubber						
Ammonia	21.7 ± 2.0	11.9 ± 6.6	45 ± 30	24 ± 13	91 ± 13	40 ± 27
Nitrous oxide	0.11 ± 0.02	0.14 ± 0.05	-26 ± 44	19 ± 15	1.2 ± 0.3	-0.3 ± 0.5
Methane	105 ± 14	108 ± 15	-4 ± 14	8 ± 5	412 ± 62	-22 ± 44
Biological air scrubber						
Ammonia	9.3 ± 1.8	1.4 ± 0.6	85 ± 6	23 ± 13	14 ± 5	12 ± 4
Nitrous oxide	0.16 ± 0.06	0.34 ± 0.09	-121 ± 64	13 ± 9	0.6 ± 0.2	-0.7 ± 0.3
Methane	72 ± 12	69 ± 11	4 ± 9	5 ± 3	105 ± 22	2 ± 9
Two-stage air scrubber						
Ammonia	19.1 ± 2.2	7.2 ± 0.9	62 ± 6	9 ± 3	22 ± 1	13 ± 1
Nitrous oxide	0.17 ± 0.04	0.49 ± 0.06	-203 ± 69	10 ± 3	0.5 ± 0.1	-1.0 ± 0.2
Methane	118 ± 22	112 ± 22	5 ± 13	12 ± 4	129 ± 21	5 ± 12

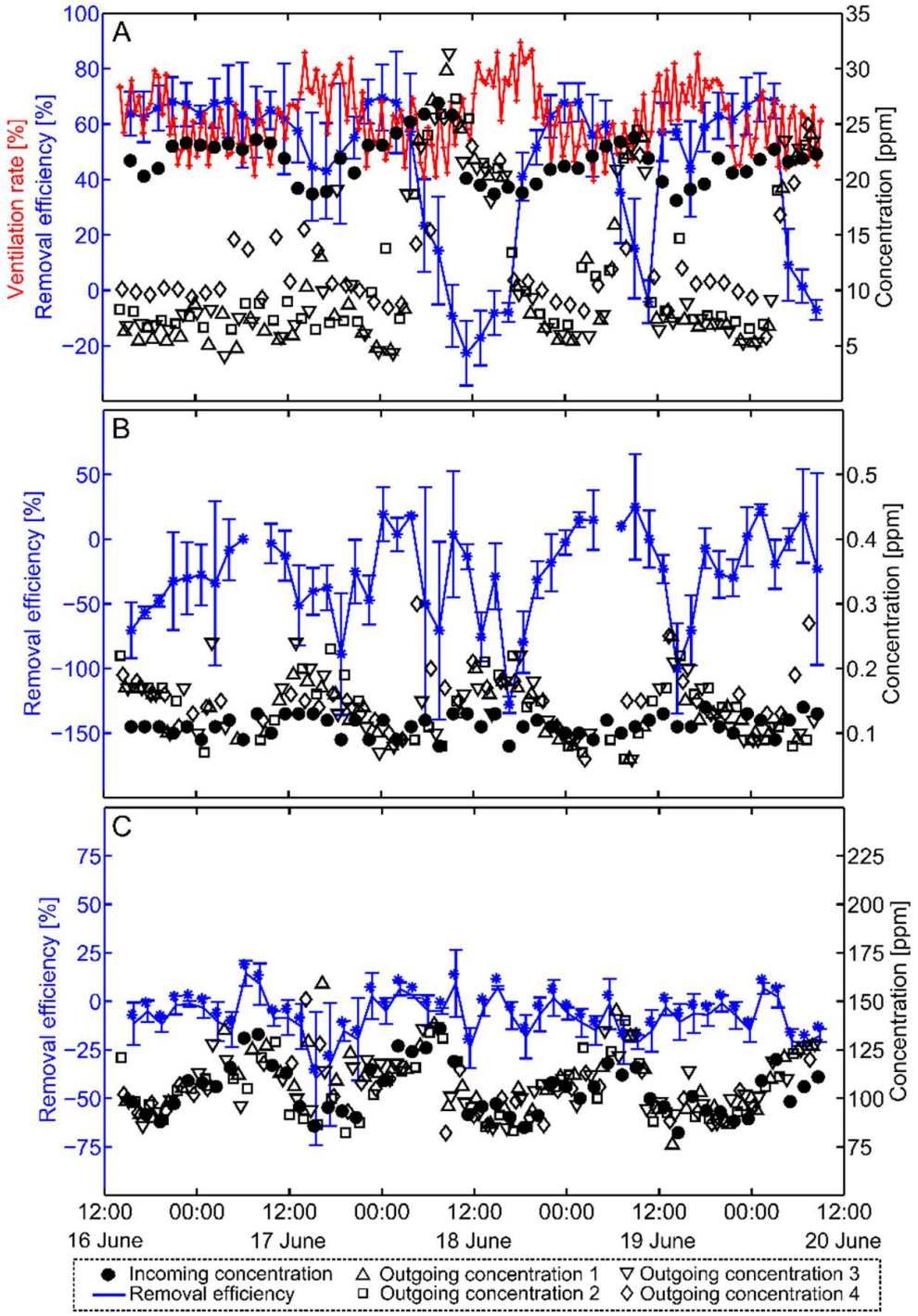

Figure 3.4. Ammonia (A), nitrous oxide (B) and methane (C) removal efficiency (blue, with standard deviation error bars) in a chemical air scrubber based on incoming and outgoing ammonia concentrations (black), and the ventilation rate (% of total ventilation; red)

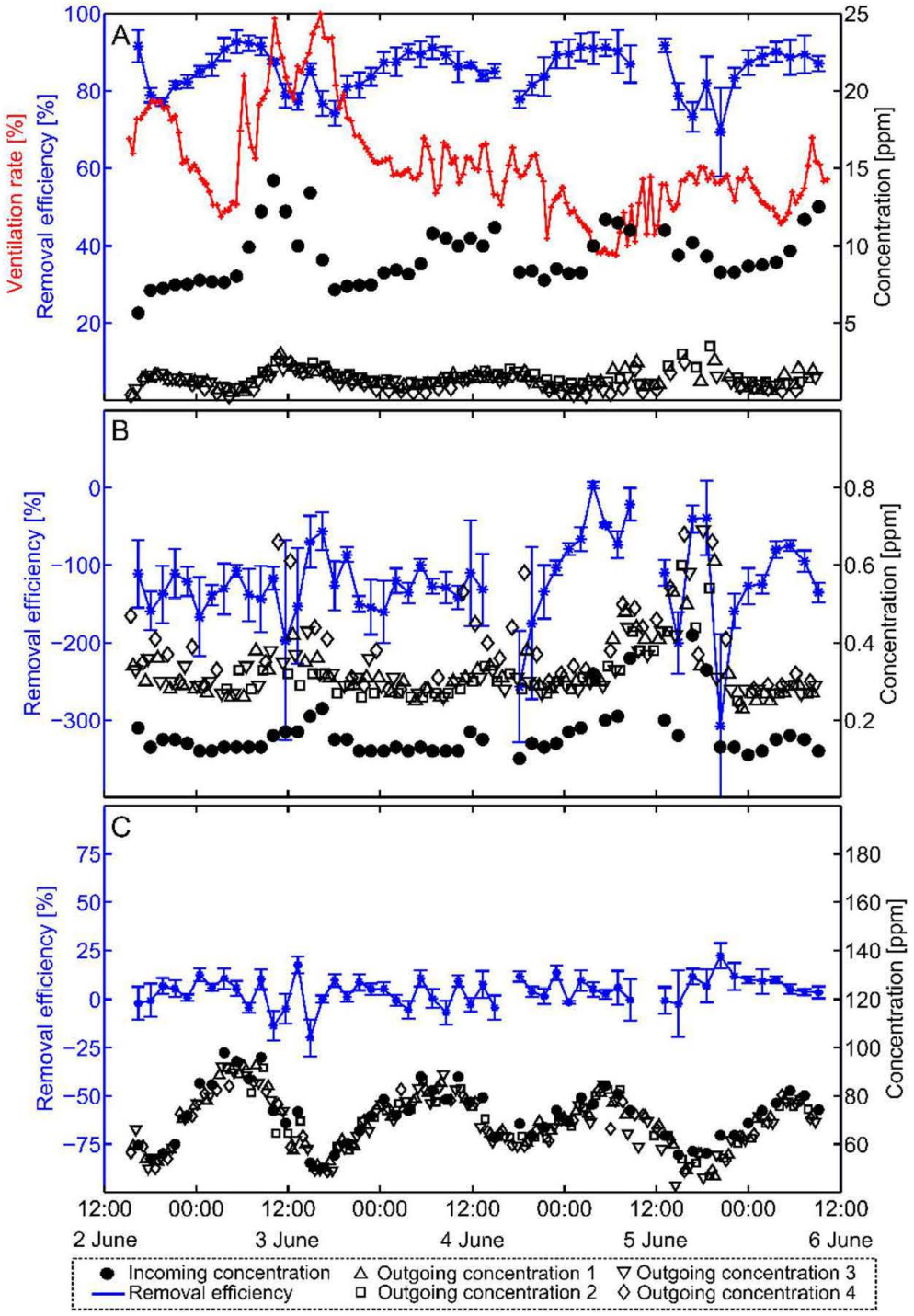

Figure 3.5. Ammonia (A), nitrous oxide (B) and methane (C) removal efficiency (blue, with standard deviation error bars) in a biological air scrubber based on incoming and outgoing ammonia concentrations (black), and the ventilation rate (% of total ventilation; red)

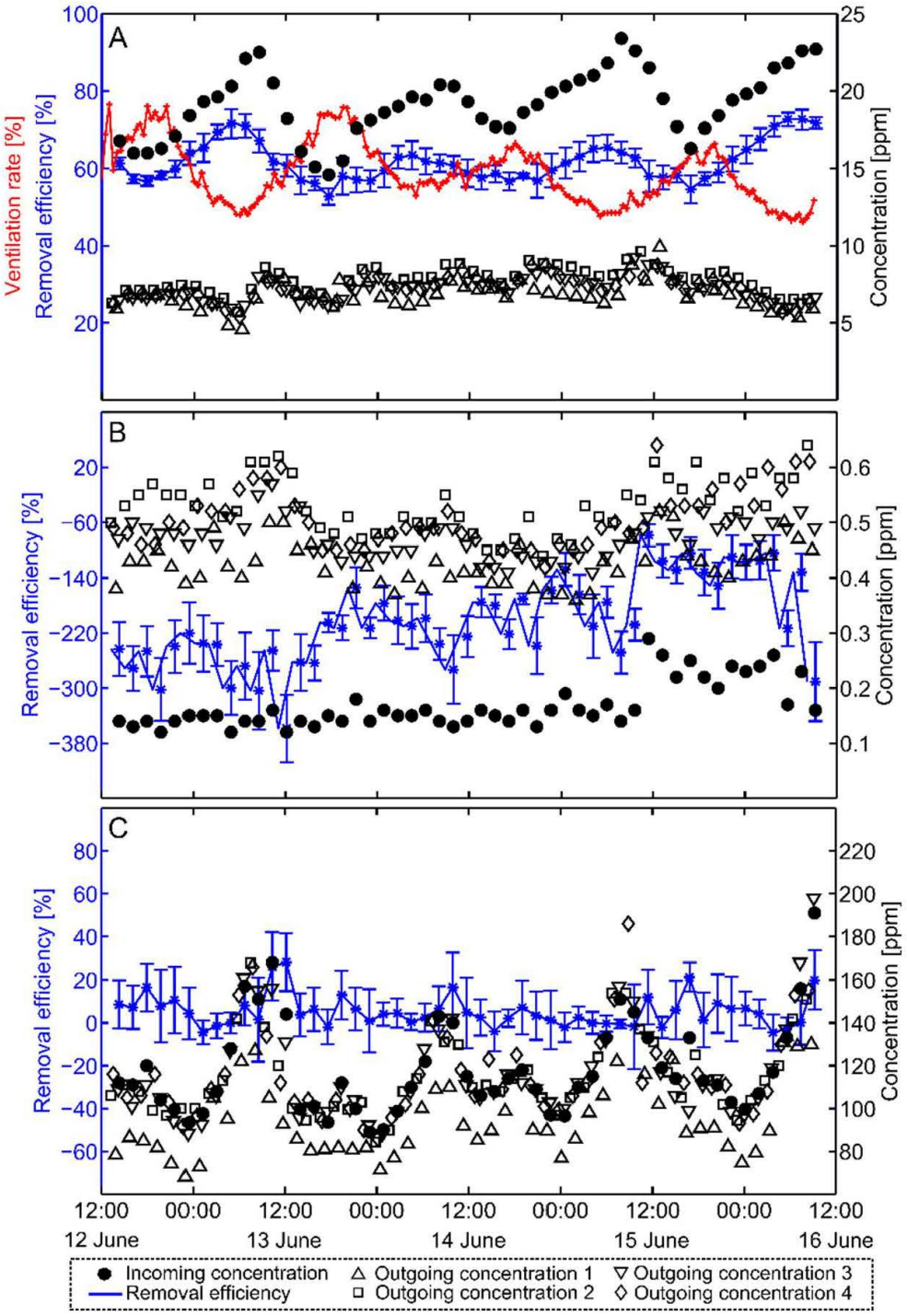

Figure 3.6. Ammonia (A), nitrous oxide (B) and methane (C) removal efficiency (blue, with standard deviation error bars) in a two-stage air scrubber based on incoming and outgoing ammonia concentrations (black), and the ventilation rate (% of total ventilation; red)

3.4.2 Washing water analysis

Table 3.4 gives an overview of the washing water analysis for the three air scrubbers. In the chemical air scrubber, a pH of 3.2 and 6.8 was found at different time instances. This also influenced the NH_4^+/SO_4^{2-} ratio ($mol.mol^{-1}$) as it was 2.3 for pH 3.2 and 1.4 to 1.9 at pH 6.8, respectively. The total nitrogen content in the washing water varied between 16.25 and 24.21 $gN.L^{-1}$.

Analysis of the washing water in the biological air scrubber showed that the total nitrogen concentration mounted on average 4.8 $gN.L^{-1}$, with values between 4.2 and 5.4 $gN.L^{-1}$. This is high compared to the standard of 3.2 $gN.L^{-1}$, set in the Flemish regulation (MB31/05/2011). Despite these high nitrogen levels, a low nitrite percentage to the total nitrogen content (between 1.0 and 2.9%) and a high nitrate percentage (between 49.5 and 68.2%) were reached. The ammonium accumulation or the percentage of ammonia to the total nitrogen content, measured between 30.8 and 48.1%. The $(NO_2+NO_3)/NH_4$ ratio was between 1.08 and 1.15 with one higher value of 2.25 measured in the last taken sample.

The washing water analysis of the two-stage air scrubber revealed that the total nitrogen concentration in the biological stage amounted on average to 2.13 $gN.L^{-1}$, with values below the maximum allowed value of 3.2 $gN.L^{-1}$ (MB31/05/2011). This water was recirculated to the preceding water stage were a total nitrogen concentration of 6.6 $gN.L^{-1}$ was reached. The washing water of the biological section showed a high nitrite accumulation (between 37.4 and 42.4%; 31.0% in water stage) and a low nitrate percentage (between 1.7 and 1.9%; 1.8% in the water stage). A high ammonium accumulation was observed, between 55.7 and 60.9% in the biological stage and 67.2% in the water stage. Thus relatively more ammonium could be found in the water stage. The $(NO_2+NO_3)/NH_4$ ratio varied between 0.64 and 0.79 in the biological stage and measured 0.49 in the water stage.

Table 3.4. Washing water analysing results for the different air scrubbers.

Date	Type	pH	pH	EC	EC	SO_4	NO_2	NO_3	NH_4
	(1)	analysis	pc	analysis	pc				
		[-]	[-]	[$mS.cm^{-1}$]	[$mS.cm^{-1}$]	[$mg.L^{-1}$]	[$mg.L^{-1}$]	[$mg.L^{-1}$]	[$mg.L^{-1}$]
16/06/2015 14:15	C	3.19	4.0	100.9	98	49240	-	-	20888
18/06/2015 10:00	C	6.80		95.1		81067	-	-	21462
18/06/2015 20:00	C	-	4.0	-	122	-	-	-	-
20/06/2015 09:30	C	6.79		101.6		88067	-	-	31124
02/06/2015 12:15	B	6.71	6.9	20.0	22	-	496	11588	3234
02/06/2015 16:00	B	6.41	6.9	24.8	22	-	504	11811	3361
05/06/2015 09:00	B	6.53	6.9	17.3	18	-	305	10254	2655
05/06/2015 16:00	B	6.22	6.9	18.1	19	-	336	10548	2771
06/06/2015 10:30	B	6.26	-	27.2	-	-	143	12792	1677
11/06/2015 12:30	T	6.91		18.2		-	2559	164	1554
12/06/2015 12:25	T*	7.18	7.1	24.5	17	-	3226	199	2057
12/06/2015 12:25	T	7.47	-	47.8	-	-	6750	517	5719
15/06/2015 16:30	T	7.20	7.4	17.3	20	-	2419	144	1244

* Type of air scrubber:
C = Chemical air scrubber
B = Biological air scrubber
T = Two-stage air scrubber
** Analysis of the water stage instead of the biological stage

3.4.3 Ventilation rate

The ventilation rate profile of the pig housing facilities is shown in Figure 3.4A, Figure 3.5A and Figure 3.6A. A daily trend can be noted with higher ventilation rates during the day of up to 80% of the maximum ventilation rate, compared to only 40% during the night for both the chemical and the two-stage air scrubber. A more irregular profile was found at the housing facility of the biological air scrubber, with only one day with high ventilation rates during the day reaching 100% of the maximum ventilation rate (June 3th), followed by a more constant profile during the remaining measuring period.

3.4.4 Air temperature and relative humidity

Figure 3.7 and Figure 3.8 show the temperature and relative humidity profile. The average incoming air temperature at the three facilities measured between 22.8°C and 24.6°C, and was up to 5°C higher during the day, when the animal activity and the outside air temperature were higher. The air temperature decreased on average 5.8°C when flowing through the air scrubber, resulting in an average outgoing air temperature of 18.1°C over the three air scrubbers, close to the wet-bulb temperature. The relative humidity increased in the air scrubbers from an average of 55.2% before the air scrubber to an average of 93.0% behind the air scrubbers. Mostly, the incoming relative humidity showed the opposite effect as the incoming air temperature with a lower relative humidity during the day. At some days during noon, the outgoing relative humidity decreased up to 61.3%.

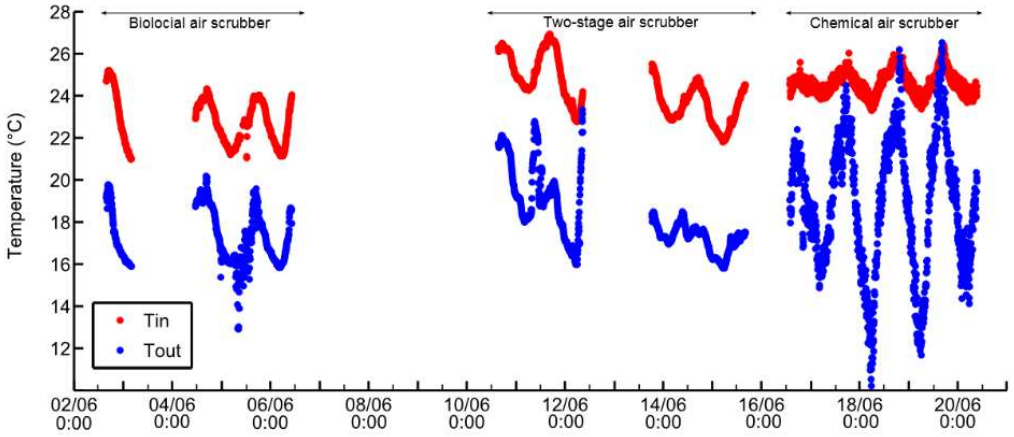

Figure 3.7. Temperature of the incoming and outgoing air at the biological, two-stage and chemical air scrubber.

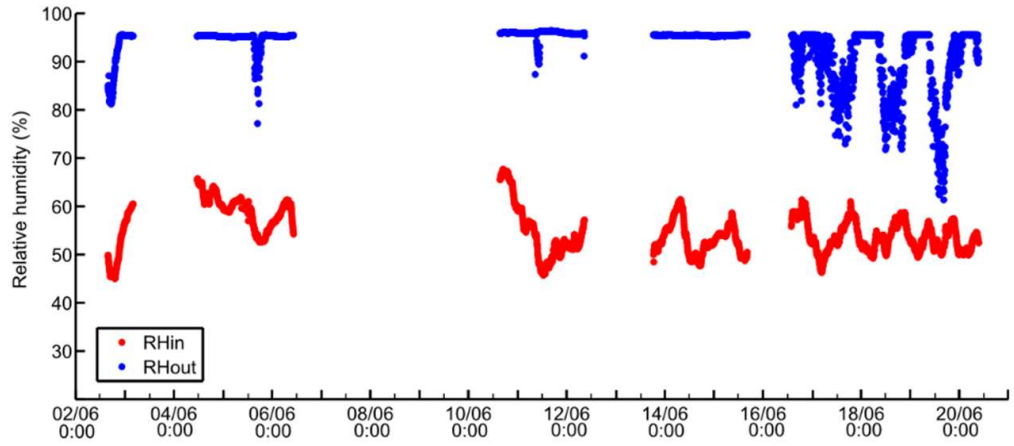

Figure 3.8. Relative humidity of the incoming and outgoing air at the biological, two-stage and chemical air scrubber.

3.5 Discussion

3.5.1 Ammonia removal efficiency

3.5.1.1 *Homogeneity over the scrubber outlet surface*

The indicative measurements with gas detection tubes and the electrochemical sensor, indicated that the two checked air scrubbers were homogeneous as the relative standard deviation of the ammonia concentration between the different measurement points was less than 30%. The homogeneity check was repeated using the continuous data confirming the homogeneity of the three air

scrubbers as the average RSD for all measured gases was below 30% (Table 3.3). However, it can be seen in Figure 3.4 that the outgoing ammonia concentrations of the chemical air scrubber showed relatively large variations (average standard deviation SD over time of 2.5 ppm) over the different sampling positions resulting in relatively large deviations in the removal efficiency. These deviations could be caused by local ammonium sulphate particles accumulating in the scrubber which could clog the air scrubber and lead to preferential ducts for the air in the packing. The biological air scrubber has on the other hand a quite large average RSD of 23%, but has only an average standard deviation over the different measurement points of 0.29 ppm. Due to the low average outgoing ammonia concentration of 1.3 ppm, this small deviation had a larger impact on the calculation of the RSD than at a larger average outgoing ammonia concentration for the chemical scrubber (11.9 ppm). Therefore, at such low outgoing ammonia concentrations the risk of wrongfully concluding that the air scrubber is not homogeneous increases. The RSD of the two-stage air scrubber is the lowest with on average 8.5%. It has a low average standard deviation of 0.61 ppm over the different measurement points and a relative high average outgoing concentration of 7.2 ppm. In general, it can thus be confirmed that the three air scrubbers were homogeneous. When using continuous data, it can also be interesting to check the average standard deviation over the different measurement points next to the RSD, as a low average outgoing concentration will negatively influence the RSD.

3.5.1.2 *Time-dependent trends*

The ventilation rate is one of the main factors causing daily trends in the outgoing ammonia concentration profile of animal housing facilities (Philippe, Cabaraux, & Nicks, 2011) and thus in the incoming ammonia concentration profile of air scrubbers. In fattening pig housing facilities, the ventilation rate shows a daily pattern with a higher ventilation rate during the day to remove the excess heat (CIGR, 1999), having a dilution effect on the indoor gaseous concentrations (K. Y. Kim et al., 2007). The incoming ammonia concentration

in the chemical and two-stage air scrubber clearly shows this diurnal variation (Figure 3.4A and Figure 3.6A). The ventilation rate of the sows and piglet housing facility shows a less profound daily trend (Figure 3.5A). When the ventilation rate increased at day 2 (3 June), an increased incoming ammonia concentration could be observed. This could be attributed to a higher animal activity at feeding time, possibly increasing the excretion of urea and faeces, by which the ammonia concentration in the pig house will increase (Blanes-Vidal, Hansen, Pedersen, & Rom, 2008; Philippe et al., 2011). Higher ventilation rates could also cause higher air velocities near manure and slurry surfaces, thus enhancing evaporation of ammonia (Groot Koerkamp et al., 1998). Furthermore, a reduced removal efficiency could be noted at an increased ventilation rate. In the chemical air scrubber, this can be seen on day 2 (17 June) when the pH was 4.0 (Figure 3.4A). This effect is also clearly visible in the two-stage air scrubber (Figure 3.6A). An increased ventilation rate will reduce the contact time for mass transfer in the air scrubber, decreasing the removal efficiency (Van der Heyden, Vanthillo, Pieters, Demeyer, & Volcke, 2016). This dependency on ventilation rate suggests that during the day, or in the summer, when the ventilation rate is the highest and the contact time will be the lowest, the ammonia removal efficiency will be lower compared to night time and in colder (winter) conditions respectively.

In general, a large variation was observed in ammonia removal efficiency of the chemical air scrubber. Even periods with a negative removal efficiency, or thus stripping of ammonia, were observed. This can be attributed to the large influence of the pH of the washing water, which varied between 3.19 and 6.80 (Table 3.4). For day 3 (18 June) the pH was 6.80 and no ammonia was removed. Although the applied discharge strategy of letting the pH increase before discharge is favourable for safety reasons as a non-acid end-product is achieved, this strategy is not favourable for the environment and should be avoided. When the pH of the washing water was below 4 (16 June), the removal efficiency for this air scrubber still only was around 60%, not reaching the required 70%. The corresponding washing water analysis shows that the total nitrogen content in the washing water stayed far below the maximum allowed content of 58.6 $gN.L^{-1}$ (MB31/05/2011). Still ammonium sulphate salt

formation was visible in the surroundings of the air scrubber and the farmer complained about regular clogging of the packing. An explanation of the low performance could lie in a possible by-pass effect whereby preferential air ducts are created inside the packing due to ammonium sulphate salt formation. Another possibility can be found in the design of the scrubber as the minimal empty bed residence time (EBRT) was 0.35 s which is rather low (Van der Heyden, Demeyer, & Volcke, 2015) and also the packing material had a low specific surface area of 80 $m^2.m^{-3}$. The loading rate per volume of packing material for this air scrubber was high compared to the other two air scrubbers. The ammonia removal efficiency of the biological air scrubber exceeded the required 70%. This could even be higher because the outgoing ammonia concentration was lower than 2 ppm and these low concentrations are difficult to measure with the photoacoustic gas monitor (Mosquera et al., 2014; Osada et al., 1998). The ammonia loading rate was quite low (Table 3.1) and analysis of the washing water shows constant high total nitrogen contents, high nitrate and low nitrite concentrations (Table 3.4), which indicate that the nitrifying bacteria were not inhibited. The extra nitrification tank containing nitrifying bacteria in granules thus had a positive influence on the operation of the scrubber, reducing the amount of discharge water and maintaining a good performance.

Despite the additional scrubber stage, the two-stage air scrubber did not reach the required 70% removal. The washing water analysis show that the nitrifying bacteria, especially the nitrite oxidizing bacteria (NOB), were inhibited as the nitrite concentrations were high and the nitrate concentration only reached 1.9% (Table 3.4), indicating an incomplete nitrification. However, the nitrogen content in the washing water of the biological stage remained below the maximum allowed content of 3.2 $gN.L^{-1}$ (MB31/05/2011). Other studies also found nitrite accumulation in biological air scrubbers and NOB were not always present (Juhler et al., 2009; Melse et al., 2011; Ottosen et al., 2011). Ottosen et al. (2011) made an overview of different studies on the effect of inhibition which shows that AOB and NOB react differently on free ammonia (FA) and free nitrous acid (FNA) but also having a different effect on the

activity and growth. It is thus not as straightforward as assumed in the Flemish regulation that inhibition will be prevented as the total nitrogen content stays below 3.2 $gN.L^{-1}$ (MB31/05/2011). Once inhibition is induced, the nitrite concentrations will increase, resulting in even further inhibition of the bacteria. Maximum discharge is then recommended in order to reduce the FA and FNA concentrations. However, this will increase the operational cost as the amount of discharge water increases.

3.5.2 Nitrous oxide removal efficiency

The incoming nitrous oxide concentrations are low, especially at the chemical air scrubber, which is at the level of the measuring limit of the photoacoustic gas monitor. Other studies with single measurements using gas chromatography (GC) also show low incoming nitrous oxide concentration levels comparable with our results (Mosquera et al., 2011). The slight increase in outgoing nitrous oxide concentration at an increased ventilation rate, hereby also increasing the variation of the outgoing concentrations, could also be attributed to the noise of the measurement itself.

In the chemical air scrubber, the incoming and outgoing nitrous oxide concentrations remained the same, as expected. In the biological and the two-stage air scrubber on the other hand, a significant production of nitrous oxide was noted as the outgoing concentration is more than double the incoming concentration. This production was due to the nitrification/denitrification reactions that take place in this type of air scrubbers. From wastewater treatment plants, it is known that there are three important formation mechanism under aerobic conditions, namely the nitrifier denitrification, the hydroxylamine oxidation and the heterotrophic denitrification (Wunderlin, Mohn, Joss, Emmenegger, & Siegrist, 2012). The most important operational parameters leading to nitrous oxide emission are: low dissolved oxygen levels, high nitrite concentrations, rapidly changing conditions in load and low COD to N content (Kampschreur et al., 2009). These are all conditions that normally prevail in biological treatment stages of air scrubbers at pig house facilities. The nitrite concentrations in the washing water of the two-stage scrubber were

much higher than the nitrate concentration. However, this was not the case for the biological air scrubber. Although it is not expected that denitrification is possible in regular air scrubbers because of the aerobic atmosphere, other literature shows that a significant amount of the present bacteria are denitrifiers, meaning that anaerobic or anoxic conditions prevail (L. Yang, Kent, et al., 2014). Both in the biological and the two-stage air scrubber, around 2% of the incoming NH_3-N was converted into N_2O-N. These levels are comparable to other studies on wastewater treatment plants (Daelman et al., 2013), biological air scrubbers (Melse et al., 2011; Melse, Ploegaert, et al., 2012) and are low compared to air scrubbers with an extra denitrification step (17, 66 and 24%) of incoming nitrogen (Melse & Mosquera, 2014). Further research is necessary to provide solutions in preventing significant nitrous oxide production to allow a more sustainable operation of these systems.

3.5.3 Fate of methane

The trend for the incoming methane concentration is the same as for ammonia. The dilution effect due to the ventilation rate can also be seen in Figure 3.4C, Figure 3.5C and Figure 3.6C: an increased ventilation rate in the morning resulted in decreased methane concentrations. During the evening, when the ventilation rate decreased, the methane concentrations increased. For the biological air scrubber in Figure 3.5C, the same decreasing and increasing trend is visible, but the daily variation in ventilation rate was less profound from day 3 on (June 4th) and can thus not completely be attributed to this influencing factor.

Methane could not be removed in any of the air scrubbers as methane is a hydrophobic component having an Henry coefficient of 0.001 mol.kg^{-1}.bar^{-1} (Lide and Frederikse, 1995). That methane cannot be removed in air scrubbers was also found in other studies using GC analysis (Melse et al., 2011; Melse, van Hattum, et al., 2012; Mosquera et al., 2011). It could be possible to introduce methane oxidising bacteria in the biological air scrubber, but the residence time should than be increased drastically to several minutes instead of seconds (Melse & Van der Werf, 2005). This could be done through an innovative recycling strategy as investigated by Estrada et al. (2014), but this

will increase the cost and is not yet realistic to apply for air scrubbers at pig housing facilities. Still profound research within this respect is needed.

3.5.4 Air temperature and relative humidity

The incoming temperature has an influence on the ventilation rate inside the pig housing facility and can thus indirect effect the air scrubber performances due to the changing ventilation rates (Van der Heyden et al., 2015). The incoming relative humidity will have an effect on the fresh water consumption because of evaporation. The decrease in temperature and increase in relative humidity inside the air scrubber are typical phenomena in air scrubbers because of sensitive and latent heat exchange (Van der Heyden, Vanthillo, et al., 2016) and were also found by other studies (Aarnink et al., 2007; Melse & Ploegaert, 2011; Mosquera et al., 2011). Variations inside the animal house depend mainly on external humidity, wall insulation type and only a small negligible effect of stocking density (Banhazi et al., 2008). For the chemical air scrubber, the highest variation in the outgoing temperature and relative humidity could be noted (Figure 3.7 and Figure 3.8). This was probably because the measurement point was placed further away from the packing material for practical reasons, possibly leading to higher influences of wind and sun radiation.

3.6 Conclusions

- The performance of three different types of air scrubbers at pig housing facilities was investigated on-line using continuous measurements for ammonia, nitrous oxide and methane, which allowed to follow the diurnal dynamics and to relate the removal efficiency pattern with underlying processes. Overall, for increased ventilation rates, the incoming concentrations of all measured pollutants were lower. Higher ventilation rates also lead to a decreased contact time in the scrubber, causing lower ammonia removal efficiencies as well. The outgoing nitrous oxide concentration showed a small increase at increased ventilation rates but since the measurement was near the detection limit, it cannot be stated if

this was due to the increased ventilation rate or due to the noise on the measurement itself.

- The ammonia removal efficiency was negatively affected by too high pH values in the chemical air scrubber and by nitrifying bacteria inhibition in the two-stage biological air scrubber. The biological air scrubber with separate nitrification tank performed well with a removal efficiency of 85 ± 6%.
- Nitrous oxide remained unaffected in the chemical air scrubber. However, it was produced in both the biological and the two-stage biological air scrubber, due to the nitrification/denitrification reactions, showing a two- and threefold increase compared to the incoming concentrations, respectively. Around 2% of the incoming ammonia was converted to nitrous oxide, which is comparable to wastewater treatment plants and low compared to air scrubbers with a denitrification step. Methane was not removed in any of the scrubbers under study, which is the result of the low water solubility of this gas. To stimulate the removal of the greenhouse gases N_2O and CH_4 in air scrubbers, further research is required.
- The homogeneity of the air scrubbers, normally checked by gas detection tubes, was investigated in more detail using the continuous data. It is advisable to check the homogeneity through the average standard deviation over the different measurement points. The interpretation of results expressed in terms of the relative standard deviation (RSD) is not straightforward since low average outgoing concentrations lead to high relative standard deviations.

4

Mechanistic modelling of a chemical air scrubber

4.1 Abstract

Chemical air scrubbers reduce the concentration of water soluble components such as ammonia from the outgoing ventilation air through absorption in water, followed by chemical conversions and removal of the end products. A mechanistic model for a countercurrent air scrubber was set up. Mass balances for ammonia, hydrogen sulphide, nitrous oxide and methane were implemented, as well as the water mass balance and heat balances. The model was validated against experimental data from a conventional fattening pig housing facility. The effect of influent characteristics, design parameters and control handles on the removal efficiency, the temperature profile and the water evaporation rate were investigated through simulation. The model was able to describe the behavior of a countercurrent chemical air scrubber.

Van der Heyden, C., Vanthillo, B., Pieters, J., Demeyer, P. & Volcke, E.I.P. (2016). Mechanistic modelling of pollutant removal, temperature and evaporation in chemical air scrubbers. Chemical Engineering & Technology 39 (10), 1785-1796. DOI: 10.1002/ceat.201500664.

4.2 Introduction

Chemical air scrubbers are widely applied for air pollution control. They are used to efficiently remove pollutants from the gas stream through absorption, typically in water, followed by chemical conversions and removal of the end products. The working principle of this type of air scrubbers has already been extensively discussed in literature by other authors (Schnelle & Brown, 2002a; Srivastava & Jozewicz, 2001; Van der Heyden et al., 2015). Applications comprise exhaust streams containing SO_2 and NO from combustion plants (Chien, Chu, & Hsueh, 2003; Thomas, Colle, & Vanderschuren, 2003), chlorine gas streams (Roy & Rochelle, 2004) or streams from agricultural applications containing mainly ammonia (BREF, 2017; Hadlocon, Zhao, et al., 2014; Hahne & Vorlop, 2001; Melse & Ogink, 2005). Optimal reactive absorption can be achieved by correct process design which is mainly dependent on proper packing selection and thorough understanding of process behaviour. For the latter purpose, the application of reliable and adequate process models is widespread (Kenig, Kucka, & Górak, 2003). Useful models can be found in literature for many chemical air scrubber applications, such as the desulphurization process (Bashipour, Khorasani, & Rahimi, 2015; Brettschneider, Thiele, Faber, Thielert, & Wozny, 2004; Kiil, Michelsen, & Dam-johansen, 1998), the absorption of odorous compounds (Meyer, Hendou, & Prevost, 1995; Overcamp, 1999) and the absorption of ammonia in water using different scrubbing systems (Huang, 2005; Khakharia et al., 2014; Ocfemia, Zhang, & Tan, 2005; Perez-Blanco, 1988; Shanableh & Imteaz, 2010; Yoon, Lim, & Chung, 2008). However, none of these models consider pollutant removal efficiency, simultaneously with temperature and water evaporation, which are essential to assess the interaction between the scrubber performance, temperature dynamics and water consumption.

The objective of this work was to set up a mechanistic model capable of predicting pollutant removal efficiency, air temperature and relative humidity distribution over the packed bed. The mass transfer of chemical species was taken into account, as well as heat transfer considering both sensible and latent heat exchange. Additionally, various correlations to calculate the mass transfer

coefficient, which is the most important parameter in absorption models, were investigated in more detail.

The model was validated using the experimental data of a chemical countercurrent air scrubber installed at a conventional fattening pig housing facility for ammonia removal, air temperature and relative humidity distribution. A case study was defined, considering a chemical air scrubber at a conventional fattening pig housing facility with 1000 animal places. In the first place, typical prevailing parameters and variables of the case study were used to simulate the (steady state) reference case. Subsequently, the effects of the typical variations in ventilation rate, incoming pollutant concentrations (loads), air temperature and relative humidity on the removal efficiency, temperature profile over the packing and evaporation rate were studied. The effects of important design parameters of air scrubbers as the packing characteristics and the packing dimensions, as well as important operational variables in air scrubbers, such as the pH of the washing water and the liquid flow rate were assessed as well.

4.3 Model description

4.3.1 Model set-up

The mechanistic model (Figure 4.1) was developed for a countercurrent chemical air scrubber and was implemented in Matlab-Simulink®. The packing material was represented by a sufficiently high number of horizontal cells consisting of an air and liquid phase (nY = 10), separated by a stagnant boundary layer at the interface (two film theory) and each of which were represented by ideally mixed continuous stirred-tank reactors (CSTR) placed in series. A fixed-step discrete solver was used with a time step chosen smaller than the refreshment rate of each cell. In each horizontal layer, a homogeneous distribution of the incoming ventilation air and spraying water is assumed, meaning that no concentration nor temperature gradients were considered along the horizontal axis. This results in a one-dimensional model, reducing complexity and calculation time. The liquid buffer tank was modelled as a separate ideally mixed reactor cell. The liquid phase of the top cell is fed by

the incoming washing liquid from the buffer tank, distributed in practice by the spray nozzles. The gas phase of the top cell contains the outgoing clean air, flowing out from the air scrubber. It was assumed that no mass transfer between the gas and liquid phases took place in this cell. The bottom cell contains the incoming exhaust air and the washing liquid in the buffer tank, which is the only CSTR that has a larger liquid volume than the other cells. Also for this cell, it was assumed that there was no mass transfer between gas and liquid phases, because the prevailing specific exchange surface area is much smaller than in the packing material itself.

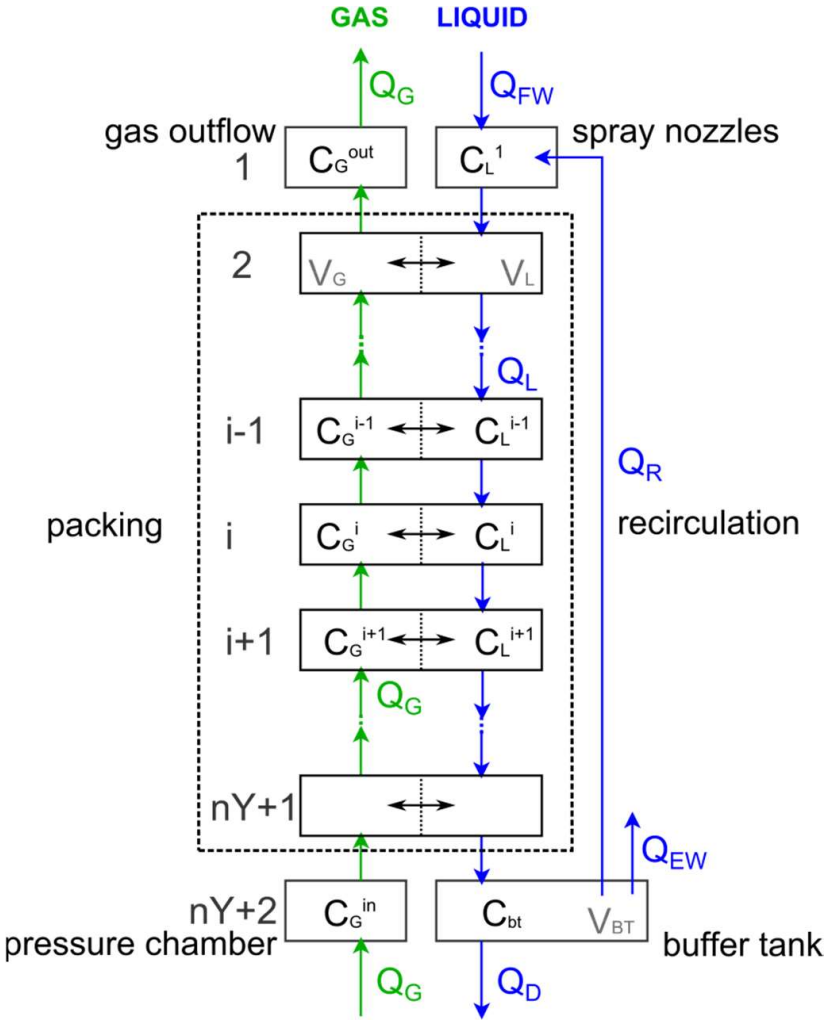

Figure 4.1. Schematic overview of the mechanistic model for a countercurrent chemical air scrubber.

4.3.2 Mass transfer

Mass transport was modeled by advection (plug-flow) in the gas and liquid bulk phases. The resistance to transfer of the pollutants between the bulk gas and the bulk liquid was considered to be entirely located in the boundary layer, beyond the boundary layer the turbulence is sufficient to eliminate concentration gradients (Froment & Bischoff, 1990). Despite the simplification (Foumeny & Pahlevanzadeh, 1990), plug-flow was assumed for both concentrations and temperature, which could be justified in air scrubbers due to the high Peclet numbers (Pe >10; Eq.4.1) and the high air velocities typically prevailing in these systems (Patankar, 1980). Dispersion or channelling phenomena were not considered in the model (Kim & Deshusses, 2003).

$$Pe = \frac{h_p \cdot u}{D} \qquad \textbf{Eq.4.1}$$

Equilibrium between the gas and the liquid phase was assumed at the interface, making Henry's law applicable. It was assumed that no reactions took place in the gas phase. Adsorption of pollutants to the inert packing material was neglected. Ionic strength could have an effect on the ammonia removal as the acidity constant and Henry coefficient depend on this parameter (Montes, Rotz, & Chaoui, 2009). However, it was not implemented in the model to reduce complexity. Liu et al. (2013) investigated the influence of ionic strength on the removal of odorous components and found this was negligible.

The individual gas and liquid phase mass balances for a pollutant (index is omitted for simplicity) in cell i were implemented as

$$\frac{dC_G^i}{dt} = \frac{Q_G}{V_{G,cell}} \cdot \left(C_G^{i+1} - C_G^i\right) - K_L^i\left[T_G^i, T_L^i\right] \cdot a \cdot \left(\frac{C_G^i}{H^i\left[T_G^i, T_L^i\right]} - C_L^i\right) \qquad \textbf{Eq.4.2}$$

$$\frac{dC_L^i}{dt} = \frac{Q_L}{V_{L,cell}} \cdot \left(C_L^{i-1} - C_L^i\right) + K_L^i\left[T_G^i, T_L^i\right] \cdot a \cdot \left(\frac{C_G^i}{H^i\left[T_G^i, T_L^i\right]} - C_L^i\right) \qquad \textbf{Eq.4.3}$$

respectively, expressing that the accumulation or depletion of a pollutant in the gas phase in cell i is due to the influx from the underlying cell with concentration C_G^{i+1} and the outflow to the overlying cell with concentration

C_G^i, as well as transfer from this pollutant from the gas phase to the liquid phase or vice versa. K_L is the total mass transfer coefficient, considering the resistances at the liquid and gas interface. The larger the K_L and the specific surface area (a), the more pollutant is removed from the gas phase. The Henry coefficient H, valid for dilute systems, expresses the ratio of the gas to liquid concentrations at the interphase surface area, considered to be in equilibrium. In the liquid phase, the total lumped concentration of a pollutant was divided into both considered species e.g. ammonia and ammonium, based on the acidity constant (K_a) and the pH of the washing water. A constant pH was assumed throughout the reactor. The temperature dependency of the Henry coefficient and acidity constant are expressed in Appendix 4A.

4.3.3 Heat transfer

The temperature distribution in the air scrubber was determined by the heat exchange in each cell due to the sensible heat exchange and the latent heat exchange due to water evaporation, assuming a perfectly adiabatic reactor. It was assumed that the heat for evaporation was in the first place provided by the gas phase while the heat for evaporation was considered to be withdrawn from the liquid phase. The heat of solution of the different pollutants was neglected as their transferred amount is relatively low. The individual gas and liquid phase energy balances in cell i thus read as

$$\frac{dT_G^i}{dt} = \frac{Q_G}{V_{G,cell}} \cdot \left(T_G^{i+1} - T_G^i\right) - h^i\left[T_G^i, T_L^i\right] \cdot a \cdot \left(T_G^i - T_L^i\right) - \frac{k_{G,H_2O}^i\left[T_G^i\right] \cdot a \cdot \left(\rho_{\text{int}}^i\left[T_G^i, T_L^i\right] - \rho_v^i\left[T_G^i\right]\right) \cdot c_{p,st} \cdot \left(T_G^i - T_L^i\right)}{\rho_G^i\left[T_G^i\right] \cdot c_{p,G}} \qquad \textbf{Eq.4.4}$$

$$\frac{dT_L^i}{dt} = \frac{Q_L}{V_{L,cell}} \cdot \left(T_L^{i-1} - T_L^i\right) + h^i\left[T_G^i, T_L^i\right] \cdot a \cdot \left(T_G^i - T_L^i\right) - \frac{k_{G,H_2O}^i\left[T_G^i\right] \cdot a \cdot \left(\rho_{\text{int}}^i\left[T_G^i\right] - \rho_v^i\left[T_G^i\right]\right) \cdot \Delta H_V^i}{\rho_L \cdot c_{p,L}} \qquad \textbf{Eq.4.5}$$

, respectively. The first term in both equations denotes the advective transport of heat between the cells, the second term the sensible heat exchange and the

third term the latent heat exchange in the cell. h is the total heat transfer coefficient [W.m^{-2}.K^{-1}], considering the resistances at the liquid and gas interface.

4.3.4 Water balance and evaporation rate

The evaporation of water involves the coupling of mass and heat transfer. The water balance was expressed in terms of the water vapour density ρ_v [kg.m^{-3}], of which the accumulation in the gas phase reads as:

$$V_{G,cell}\frac{d\rho_v^i}{dt} = Q_G \cdot \left(\rho_v^{i+1} - \rho_v^i\right) - k_{G,H_2O}^i\left[T_G^i\right] \cdot a \cdot \left(\rho_{v,\text{int}}^i\left[T_G^i, T_L^i\right] - \rho_v^i\left[T_G^i\right]\right) \quad \textbf{Eq.4.6}$$

with $k_{G,H2O}$ the mass transfer coefficient at the gas side [m.s^{-1}] and ρ_{int} the water vapour density at the interphase [kg.m^{-3}]. The first term represents the advective transport of water vapour between the cells, the second one the transfer of water vapour between the gas and liquid phase in the cell. The amount of water evaporated in each cell per time instant was negligible, allowing to assume that the volume of the liquid phase was the same for all cells along the packing material (see Appendix 4C).

The water vapour density of the incoming air (ρ_v^{in}) was calculated from the ideal gas law (Eq.4.7). Its vapour pressure (p_v^{in}, Eq.4.8) was calculated based on the incoming relative humidity (RH) and the saturated vapour pressure (p_{sv}^{in}), which is dependent on the incoming air temperature (see Appendix 4A).

$$\rho_v^{in} = \frac{p_v^{in} \cdot M_{H_2O}}{R \cdot \left(T_G^{in} + 273.15\right) \cdot 1000 g.kg^{-1}} \quad \textbf{Eq.4.7}$$

$$p_v^{in} = \frac{RH^{in}}{100} \cdot p_{sv}^{in}\left[T_G^i\right] \quad \textbf{Eq.4.8}$$

The flow rate of water evaporating was calculated based on the difference in water vapour density between the cell containing the outgoing air and the cell containing the ingoing air.

$$Q_{EW} = Q_G \cdot \frac{1}{\rho_L^1} \cdot \left(\rho_v^1 - \rho_v^{in}\right) \quad \textbf{Eq.4.9}$$

The total water consumption was not only dependent on the evaporation rate but also on the amount of discharged water (Q_D). Both need to be replaced to maintain a constant water volume in the buffer tank.

4.3.5 Mass and heat transfer coefficient calculation

The total mass transfer coefficient K_L in Eq.4.2 and Eq.4.3 is determined including both diffusion and convection, taking into account the resistance for mass transfer at the gas and the liquid boundary layers. This total mass transfer resistance is characterised by the mass transfer coefficients in gas and liquid phases (respectively k_G and k_L) and the Henry coefficient, H, according to the generally applied two-film theory (Lewis & Whitman, 1924):

$$\frac{1}{K_L^i} = \frac{1}{H^i \cdot k_G^i} + \frac{1}{k_L^i} \qquad \textbf{Eq.4.10}$$

Both gas and liquid phases can constitute significant mass transfer resistances (Ibusuki & Aneja, 1984; Lewis & Whitman, 1924; Wang, Yuan, & Yu, 2005). The Henry coefficient has a large impact on whether the mass transfer for a given contaminant is limited by the gas side resistance, the liquid side resistance or both (Andreasen, Liu, Ravn, Feilberg, & Poulsen, 2013). The transfer of highly soluble compounds (low H) is generally limited by the gas side resistance, while highly volatile compounds (high H) are mostly limited by the liquid side resistance (Roberts, Hopkins, Munz, & Riojas, 1985). Typical values for a countercurrent packed scrubber with a specific surface area between 10 and 350 $m^2.m^{-3}$ lie between $7.5 \cdot 10^{-4}$ and $5.0 \cdot 10^{-2}$ $m.s^{-1}$ for k_G and between $4 \cdot 10^{-5}$ and $2 \cdot 10^{-4}$ for k_L (Overcamp, 1999). To adequately describe both highly soluble compounds and highly volatile compounds in this model, both gas and liquid mass transfer were taken into account. Besides pollutants, the transfer of water between the gas and liquid phase was considered as well. The total convective mass transfer coefficient for water is completely determined by the mass transfer coefficient for the gas phase as the one for the liquid phase is infinitely large ($k_{L,H_2O} \approx \infty$).

The mass transfer coefficients for the gas and liquid phases ($F = L$ or G) are defined as the ratio between the diffusion coefficient (D) and the thickness of the film (y_F):

$$k_F^i = \frac{D_F^i}{y_F} \qquad \textbf{Eq.4.11}$$

As the thickness of the film is not known, empirical correlations are typically used to determine the mass transfer coefficient (Wang et al., 2005). The accuracy of these correlations usually lies around 30%, but larger errors are not uncommon (Cussler, 2009). The correlations are useful for the preliminary design of small pilot plants but should always be checked experimentally before the actual construction of a full-scale plant. To understand the behaviour of air scrubbers, the use of these correlations in the model is very convenient and can be accepted. Various correlations to calculate the gas and liquid mass transfer coefficient are available in literature. The dimensionless form of the correlations for the mass transfer coefficient may disguise the qualitative similarities between them. They are typically dependent on the Reynolds number (Re) and the Schmidt number (Sc) (Wang et al., 2005). Additionally, they are dependent on the shape and structure of the considered random or structured packing materials. The packing was further assumed to be completely wetted (no dry spots) for reasons of simplicity and generality of the model. In practice, the wetted specific surface area may be somewhat lower and can be calculated from correlations which highly depend on the specific packing characteristics (Wang et al., 2005).

Figure 4.2 compares the values of mass and heat transfer coefficients obtained with three frequently used correlations for randomly or structured packed countercurrent columns (see Appendix 4B for the equations), namely those of Onda et al. (1968), Shulman et al. (1955) and Billet and Schultes (1999) for three different types of packing material. The heat transfer coefficient was calculated from the mass transfer coefficient, using the heat and mass transfer analogy of Chilton-Colburn (1934):

$$\frac{Sh}{Nu} = \left(\frac{Sc}{\mathrm{Pr}}\right)^m \qquad \textbf{Eq.4.12}$$

with *Sh* the Sherwood number, *Nu* the Nusselt number, *Sc* the Schmidt number and *Pr* the Prandtl number. The power coefficient *m* is set at 1/3 for most configurations.

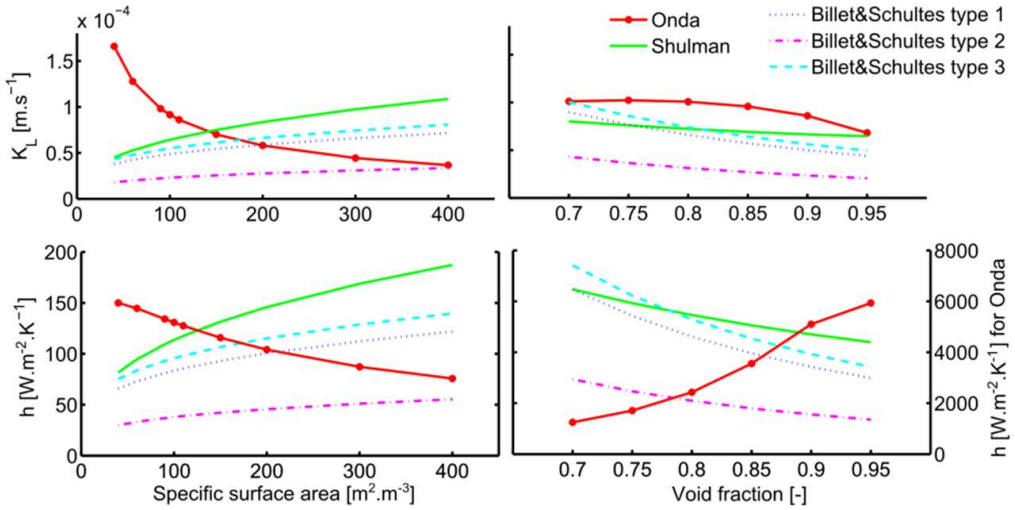

Figure 4.2. Comparison of K_L for ammonia and *h* values for the correlations of Onda, Shulman and Billet & Schultes for 3 different types of packing materials in terms of specific surface area *a* ($m^2.m^{-3}$) with a void fraction of 0.90 (left side) and void fraction of the packing material (-) with a specific surface area of 110 $m^2.m^{-3}$ (right side) . The values for c_G and c_L for type 1 (plastic dumped Pall rings of 50 mm) are 0.368 and 1.239, for type 2 (plastic Euroform) 0.167 and 0.973 and for type 3 (metal Montz packing B2-300 0.422 and 1.165, respectively.

For all shown correlations, except for Onda et al. (Onda et al., 1968), the mass and heat transfer coefficients increase with increasing specific surface area. The heat transfer coefficient calculated with correlation of Onda et al. (1968) lies around 7000 $W.m^{-2}.K^{-1}$, which is one order of magnitude higher than with the other correlations. Since the correlation of Billet and Schultes (1999) was found to be the most practical correlation as only little specific packing parameters are necessary and it is valid for different packing materials, this one was incorporated in the model:

$$k_G^i = cp_G \cdot \frac{a^{0.5} \cdot D_G^i\left[T_G^i\right]}{\sqrt{d_h\left(\varepsilon - h_L\right)}} \cdot \left(\frac{\rho_G^i\left[T_G^i\right] \cdot u_G}{a \cdot \mu_G^i\left[T_G^i\right]}\right)^{3/4} \cdot Sc\left[T_G^i\right]^{1/3} \qquad \textbf{Eq.4.13}$$

$$k_L = cp_L \cdot \left(\frac{\mu_L\left[T_L^i\right] \cdot g}{\rho_L}\right)^{1/6} \cdot \left(\frac{D_L\left[T_L^i\right]}{d_h}\right)^{0.5} \left(\frac{u_L}{a}\right)^{1/3} \qquad \textbf{Eq.4.14}$$

with d_h the hydraulic diameter of the packing material and h_L the liquid hold-up of the packing, calculated according to Billet & Schultes (1999):

$$d_h = 4\frac{\varepsilon}{a} \quad \textbf{Eq.4.15}$$

$$h_L = \left(12\frac{1}{g}\cdot\frac{\mu_L}{\rho_L}\cdot u_L\cdot a^2\right)^{1/6} \quad \textbf{Eq.4.16}$$

4.4 Case study

A chemical air scrubber was considered to treat the exhaust air from a conventional fattening pig housing facility with 1000 animal places (Table 4.1). The components considered in the gas phase were ammonia as the most important component in the exhaust air from piggery houses (Melse et al., 2009), hydrogen sulphide as a model component that can be linked with odour removal (Hansen, Adamsen, et al., 2012), nitrous oxide and methane for their greenhouse potential and water for the evaporation rate and temperature predictions. The considered incoming concentrations are based on typical values found in the exhaust air of fattening pig housing facilities. The components considered in the liquid phase were ammonia (NH_3) and ammonium (NH_4^+), lumped into total ammonia nitrogen (TAN) and taking into account the chemical equilibrium between both species, hydrogen sulphide (H_2S) and the hydrosulphide ion (HS^-), lumped into total sulphide (TS), nitrous oxide (N_2O) and methane (CH_4).

The dimensions of the considered air scrubber were based on typical minimal empty bed residence times (EBRTs) for ammonia removal in chemical air scrubbers, with an average value of 0.4 s (InfoMil, 2015; Melse & Ogink, 2005), corresponding to a minimum packing volume of 6.7 m³ for the given number of pigs and the typically applied maximum ventilation rate for design purposes of 60 $m^3.(animal.h)^{-1}$.

An acid was added to the washing water to decrease the pH which results in an increase in driving force for ammonia absorption. The pH value of the washing water was assumed to be constant, at a value of 4 for the reference case. The liquid flow rate Q_L was set to 10 $m^3.h^{-1}.m^{-2}$ cross sectional area (Van der

Heyden et al., 2015). The recirculation rate was set to 100% to reduce the water consumption. The washing water is recirculated until its ammonium concentration becomes too concentrated, which is in practice typically controlled on a time basis or through monitoring the electrical conductivity (EC) (Van der Heyden et al., 2015). A maximum ammonium concentration of 30 $gN.L^{-1}$ is mostly reached in the washing water (Van der Heyden et al., 2015) and is in the reference case assumed as set point. At discharge, one third of the buffer tank was discharged and replaced with fresh water, a commonly used practice. Initially, the liquid phase contained no pollutants.

The typical prevailing parameters and variables of the case study were used to first simulate the (steady state) reference case. Subsequently, a scenario analysis was performed, assessing the effect of the influent characteristics, important design aspects as well as important operational aspects in air scrubbers.

Table 4.1. Air scrubber design specifications and operational parameters. All values without a reference are based on practical experience. The given minimum and maximum values indicate the range wherein the effect of the parameters on the removal efficiency and evaporation rate were tested (scenario analysis).

Parameter	Symbol	Unit	Reference case		Scenario analysis			
					Min		Max	
INFLUENT CHARACTERISTICS								
Number of pigs	n_{pigs}	[animal]	1000	[A]				
Ammonia load	EF_{NH3}	[kg NH_3. (animal.y)$^{-1}$]	3	[1]	0.5	[1]	6.8	[1]
Incoming H_2S concentration	$C_{H_2S}^{in}$	[ppm]	1	[2]				
Incoming N_2O concentration	$C_{N_2O}^{in}$	[ppm]	1	[3]				
Incoming CH_4 concentration	$C_{CH_4}^{in}$	[ppm]	100	[3]				
Ventilation rate	Q_{vent}	[m^3.(animal.h)$^{-1}$]	35	[A]	6	[4]	80	[4]
Gas exhaust temperature	T_G^{in}	[°C]	22.4	[5]	15	[A]	35	[A]
Relative humidity	RH^{in}	[%]	65	[6]	45	[A]	99	[A]
DESIGN ASPECTS								
Packing height	h_P	[m]	0.90	[A]	0.3		2.5	
Packing length	l_p	[m]	3	[A]				
Packing width	w_p	[m]	2	[A]				

Parameter	Symbol	Unit	Reference case		Scenario analysis			
					Min		Max	
DESIGN ASPECTS								
Cross-sectional area	$A_{LW} = w_p \cdot l_p$	[m^2]	6	[A]	1	[A]	16.5	[A]
Buffer tank height	h_{BT}	[m]	0.30	[A]				
Specific surface area	a	[$m^2.m^{-3}$]	110	[7]	40	[A]	300	[A]
Void fraction	ε	[-]	0.936	[7]	75	[A]	99	[A]
Packing specific gas coeff.	c_G	[-]	0.167	[7]				
Packing specific liquid coeff.	c_L	[-]	0.973	[7]				
OPERATIONAL PARAMETERS								
pH washing water	pH	[-]	4	[8]	2	[A]	14	[A]
Water flow rate	Q_L	[$m^3.h^{-1}.m^{-2}$]	10	[8]	0.5	[A]	30	[A]
Recirculation fraction	f_R	[%]	100	[8]	60	[A]	100	[A]

[A] = assumed parameter values
[1] (Philippe et al., 2011)
[2] (Zhu, Jacobson, Schmidt, & Nicolai, 2000a)
[3] (Ulens et al., 2014)
[4] (Klimaatplatform Varkenshouderij, 2014)
[5] (Geers et al., 1989)
[6] (Radon et al., 2002)
[7] (Billet & Schultes, 1999)
[8] (Van der Heyden et al., 2015)

4.5 Model validation based on experimental data

Experimental data were obtained during 35 hours in June 2014 from a chemical air scrubber installed on a fattening pig housing facility for 1250 animals (Table 4.2), which is in detail explained in Chapter 3.

The model was first validated for ammonia removal, air temperature and relative humidity distribution using the experimental data of a chemical countercurrent air scrubber. The measurements for ingoing ammonia concentration, air temperature, relative humidity and ventilation rate were used as input for the simulation. The initial conditions for the simulation were set to the steady state conditions corresponding with the influent conditions at that time instant.

Table 4.2. Characteristics of the chemical air scrubber in the measuring campaign.

Parameter	Symbol	Unit	Value
Packing height	h_p	[m]	0.38
Packing length	w_p	[m]	2.25
Packing width	l_p	[m]	5.46
pH washing water	pH	[-]	3.19 (a)
EC washing water	EC	[$mS.cm^{-1}$]	100.9 (a)
Water flow rate	Q_L	[$m3.h^{-1}.m^{-2}$]	10
Recirculation fraction	f_R	[%]	100

(a) sample taken on 2014/06/16, 14.15h)

4.6 Results and discussion

4.6.1 Model validation

The measured ingoing ammonia concentration varied between 18.1 and 23.6 ppm_v, depending on the ventilation rate (Figure 4.3). For instance, at noon, when the animal activity is the highest and the indoor temperature increases, a higher ventilation rate is applied, resulting in a lower ammonia concentration entering the air scrubber. The ammonia concentration reduced in the air scrubber to an average outgoing ammonia concentration of 8.2 ± 1.2 ppm_v. The

simulated outgoing ammonia concentration amounted 7.4 ± 0.6 ppm_v and followed the experimental effluent ammonia concentration with an average of 1.2 ppm difference, within one standard deviation of the measurements.

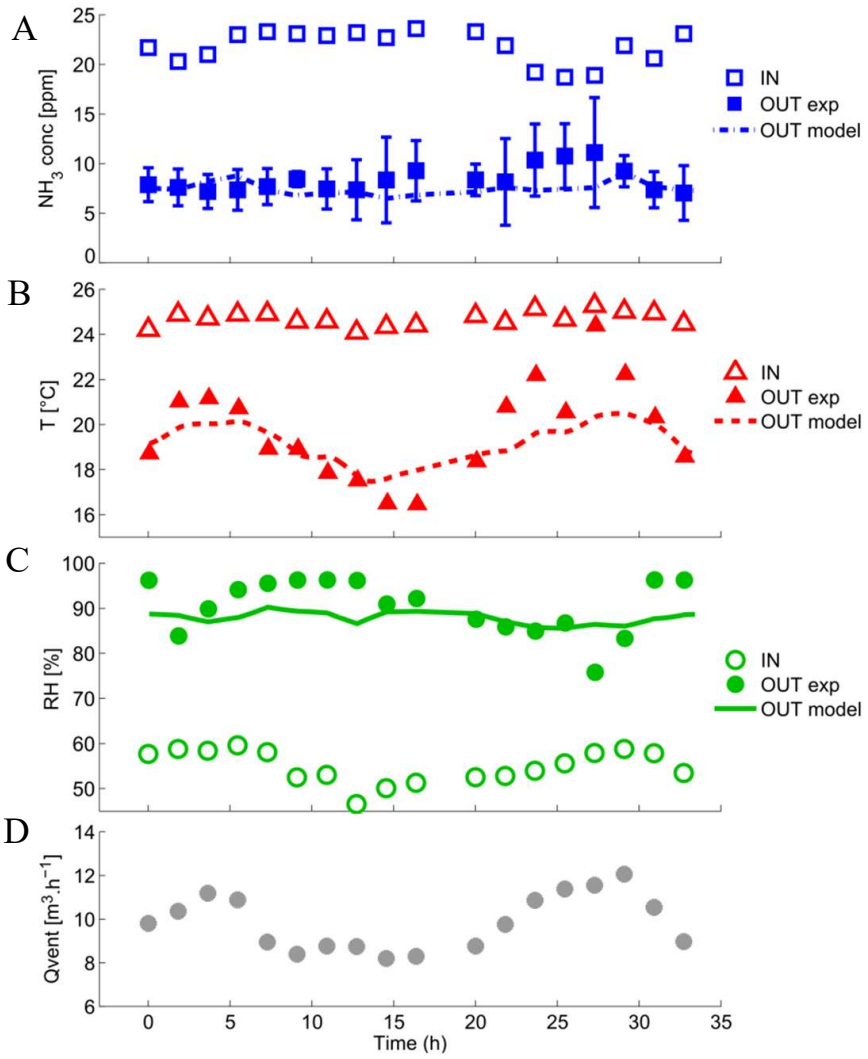

Figure 4.3. Comparison between experimental data and model simulation results for ammonia concentration (A), air temperature (B) and relative humidity (C), applying the incoming ventilation rate (D) as a simulation input.

The temperature of the incoming exhaust gas was almost constantly at 24.8 ± 0.4°C and decreased approximately 5°C throughout the scrubber to an exhaust temperature of 20.1 ± 2.5°C. Such temperature drop is typical for this type of systems (Ellen, Hol, Hoofs, & Mosquera, 2007; Melse & Willers, 2004). The simulated outgoing temperature was also 5°C lower than the influent temperature and followed the measured outgoing air temperature profile well. The outgoing temperature is higher at a higher ventilation rate because more

thermal mass enters the air scrubber. The maximum under- or overestimation was only 4.1°C, respectively, with an average difference of only 1.1°C.

The measured relative humidity changed from an incoming value of 54.2 ± 3.4% to an outgoing value of 87.1 ± 9.0%. This latter shows a large variation with values between 64.1% and 95.5%. The simulation results of the relative humidity showed a rather constant value of 87.1 ± 1.2%. In comparison, the simulated and modeled outgoing relative humidity had a 5.0% average difference, the maximum difference was 10.6%. The larger variation in the relative humidity measurements than in the simulated results can be attributed to the weather conditions, influencing the sensor which was placed outside the air scrubber. Overall, the model was well able to predict the outgoing ammonia concentration, temperature and relative humidity.

4.6.2 Reference case

The steady state profiles over the air scrubber for the reference case are displayed in Figure 4.4. The gas phase ammonia concentration decreased from 0.008 $gN.m^{-3}$ (14 ppm) at the scrubber inlet to 0.0013 $gN.m^{-3}$ (2.1 ppm) at the scrubber outlet, corresponding to a removal efficiency of 84%. The odorous component H_2S and the greenhouse gases N_2O and CH_4 were hardly removed in the air scrubber (results not shown). This holds for all components with a low solubility in water (high Henry coefficient). The relative humidity of the exhaust air gradually increased while flowing through the air scrubber from 65% to 90%, due to evaporation of the washing water. The assumption of the constant liquid cell volume in the packing regardless the evaporation of water was justified as the calculated maximum evaporation at extreme conditions (35°C and a relative humidity of 45%) only led to a decrease of the water volume in the packing of 0.26% (see Appendix 4C). Through contact with the colder washing water, the gas temperature decreased from 22.4°C to 18.0°C, close to the wet bulb temperature of 17.9°C. The liquid temperature slightly increased when flowing down through the air scrubber (Figure 4.4C). This small increase was cancelled due to evaporation in the lower part of the scrubber and the liquid equilibrium temperature of 17.1°C was reached again.

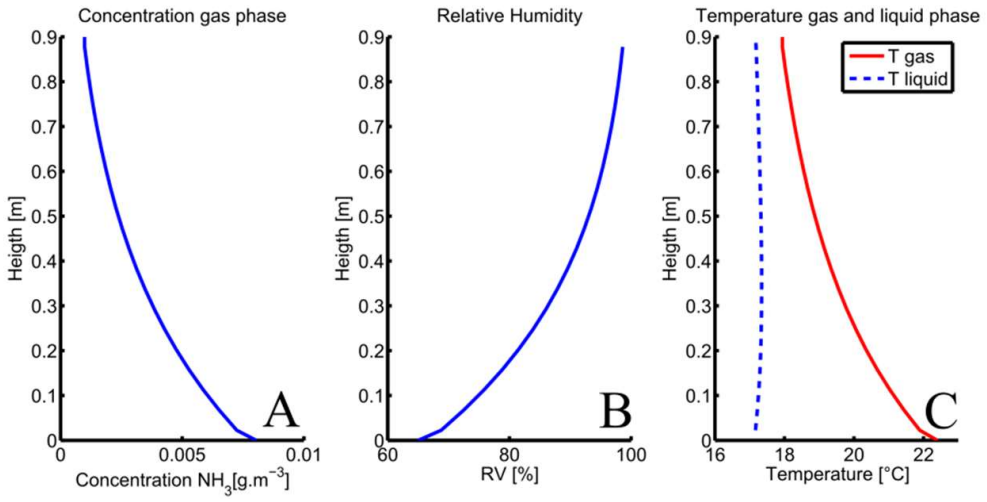

Figure 4.4. Simulated steady state profiles in the air scrubber for ammonia in gas phase (A), relative humidity (B) and the temperature of gas and liquid phase (C). The scrubber inlet and outlet correspond to position 0 and 0.9m respectively.

Despite an increasing ammonia concentration in the buffer tank (by 760 $gN.m^{-3}.d^{-1}$, results not shown), the gaseous ammonia profile inside the air scrubber and thus the removal efficiency remained constant as the driving force was still high enough. To keep the ammonia concentration in the washing water below 30 $gN.L^{-1}$, the first discharge will take place after 40 days. If one third of the water is discharged and replaced by fresh water, this means that every 13 days the threshold will be reached again. Under the constant conditions assumed in this case study, the yearly water loss due to evaporation amounted to 628 m^3, which is comparable to values found at full-scale countercurrent chemical air scrubbers (Innovatiesteunpunt, Inagro, VCM, & Provincie Vlaams-Brabant, 2014; Melse & Ogink, 2004).

4.6.3 Influence of influent characteristics

4.6.3.1 *Ventilation rate, pollutant concentration and load*

The ventilation rate shows diurnal as well as seasonal variations at animal housing facilities. Typically, high concentrations will occur during winter and at night when the ventilation rate is low and low concentrations will prevail in summer and during noon when the ventilation rate is at its maximum to remove excess heat (Hadlocon, Zhao, et al., 2014; Ulens et al., 2014). Figure 4.5A and Figure 4.5D shows the simulation result of a varying ventilation rate on the ammonia concentration in the air scrubber and the ammonia removal

efficiency, respectively. Assuming a constant emission factor, i.e. a constant incoming ammonia load, an increased ventilation rate implies a decreasing incoming ammonia concentration (Figure 4.5A). Nevertheless, the outgoing ammonia concentration increased for higher ventilation rates (Figure 4.5A), resulting in a decreasing removal efficiency (Figure 4.5D). Decreasing the incoming ammonia concentration while maintaining a constant ventilation rate did not affect the removal efficiency (Figure 4.5E), since the effluent ammonia concentration decreased accordingly to the lower incoming load (Figure 4.5B). Increasing the ventilation rate while keeping the incoming ammonia concentration constant (Figure 4.5C), led to the same decrease in removal efficiency as observed for a constant ammonia load, indicating that the decrease of the ammonia removal efficiency at higher ventilation rate was only caused by the decrease in contact time rather than by a decreasing incoming ammonia concentration (Figure 4.5F).

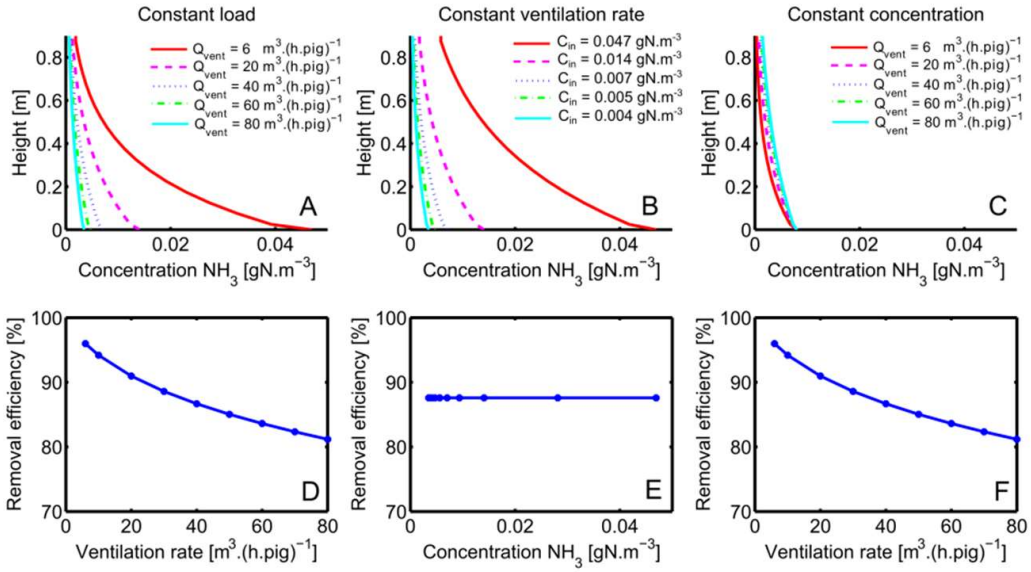

Figure 4.5. Simulated influence of ventilation rate at constant load EF_{NH3} (A, D), different incoming concentration at constant ventilation rate Q_{vent} (B, E) and different ventilation rate at constant incoming concentration $C_{in,NH3}$ (C, F) on the ammonia gas profile in air scrubber and the ammonia removal efficiency, respectively.

A higher ventilation rate also implied that more water evaporated (results not shown), because of the increased incoming thermal mass and the higher velocity resulting in a higher mass transfer coefficient. Additionally, the simulated gas temperature decreased less with higher flow rates because of

shorter contact times and the larger air mass coming into the scrubber. Through this increase in incoming thermal mass, higher liquid temperatures were modeled in the lower part of the scrubber, resulting in an increase of the temperature in the buffer tank.

4.6.3.2 *Air temperature*

The simulated influence of the air temperature on the pollutant removal efficiency was found negligible in this case (only 0.1% for a 20°C change in air temperature, results not shown). For an increasing air temperature, the gas pollutants will be less soluble (higher H) but the mass transfer will increase (higher K_L), effects that counteract each other. An air temperature increase from 20°C to 25°C results in a 20.02% higher H and a 20.11% higher K_L, illustrating the negligible overall effect. Additionally, the acidity constant of ammonia increases at increasing temperature, implying a decreased driving force. A negligible effect of temperature on the ammonia removal efficiency was also reported in tests of an acid spray scrubber (Manuzon et al., 2007) and an ammonia regeneration scrubber (Shah, Westerman, Munilla, Adcock, & Baughman, 2008). Additionally, the uptake of the heat and water balance in this study allowed to simulate the increased rate of water loss in the air scrubber, preceded by a higher condensation (negative evaporation) after start-up of the air scrubber (Figure 4.6).

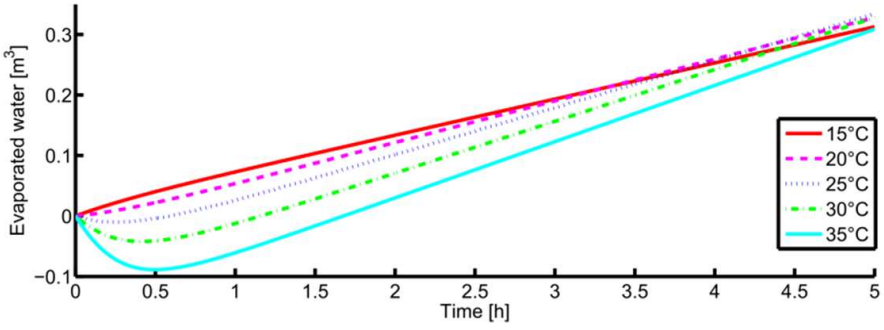

Figure 4.6. Simulated influence of air temperature on water loss in the air scrubber

4.6.3.3 *Relative humidity*

While the effect of the relative humidity on the removal efficiency of pollutants was very small (less than 1% difference between 45 and 99%, results not shown), its effect on water loss through evaporation was found significant. A

high relative humidity implied a high water vapour pressure in the incoming air and thus a lower driving force for evaporation. Given that there was less evaporation, also the latent heat transfer decreased, resulting in a smaller temperature difference between incoming and outgoing air. The equilibrium temperature of both the gas and the liquid phases inside the air scrubber was therefore higher. The water loss was much lower at higher relative humidity (Figure 4.7). A comparable linearly decreasing water loss with increasing relative humidity was found by Shah et al. (Shah et al., 2008). After start-up, a higher increase in condensation occurred at higher relative humidity because the incoming water vapour pressure was already higher than the saturated vapour pressure, meaning that the temperature first needed to increase before water was evaporated (Figure 4.7).

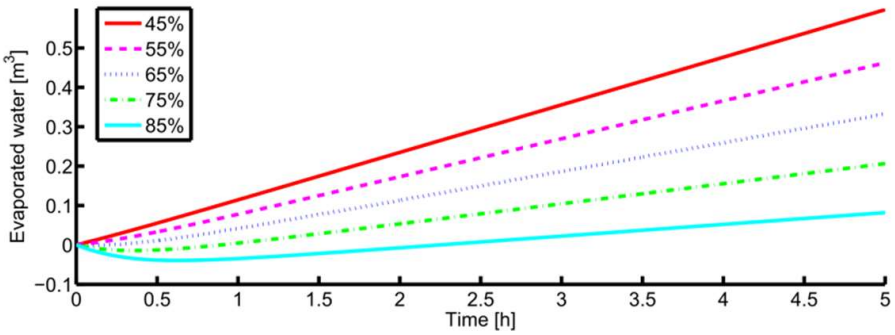

Figure 4.7. Simulated influence of the relative humidity on the water loss in the air scrubber

4.6.4 Influence of design

Using a packing material with a higher specific surface area resulted in an increasing removal efficiency (Figure 4.8) as more contact surface area was available for mass and heat transfer and the mass transfer coefficient increased (Figure 4.2). In practice however, the specific surface area cannot be increased too much to avoid the risk of clogging due to formed ammonium salts. Accumulated ammonium salts will also decrease the specific surface area during operation, implying that the removal efficiency will decrease accordingly.

Figure 4.8 displays the influence of the specific surface area and packing dimensions on the ammonia removal efficiency. Increasing the volume of the

packing material resulted in an increased removal efficiency. With respect to the effect of the cross-section, only its area influences the removal efficiency while the aspect ratio does not. Increasing the cross-sectional area of the packing resulted in a smaller positive effect than increasing its height, for the same contact area above 594 m^2 (reference case). This is explained by the decreased gas velocity associated with an increased cross-sectional area, reducing the mass transfer coefficient (Eq.4.12). It can further be noted that changing the dimensions of the air scrubber will also affect the pressure drop, which strongly influences the energy costs. Increasing the height of the air scrubber (for a constant packing volume) increases the ammonia removal efficiency, but reduces the cross-sectional area and thus increases the pressure drop; doubling the air velocity through the scrubber quadruples the pressure drop (Andreasen & Poulsen, 2013). The optimal packing height will compromise maximal removal efficiency and minimal pressure drop.

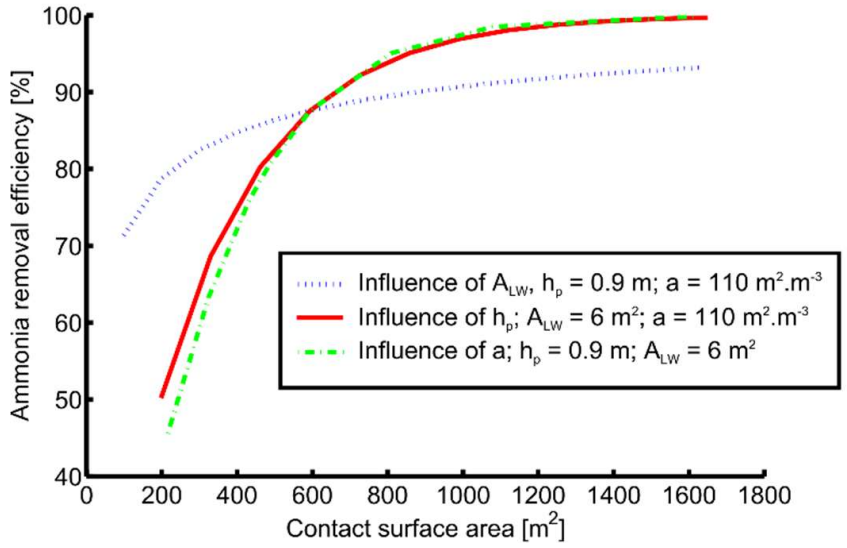

Figure 4.8. Simulated influence of the air scrubber packing dimensions (height h_p in m and cross-sectional area A_{LW} in m^2) and the specific surface area (a in $m^2.m^{-3}$) on the ammonia removal efficiency.

4.6.5 Influence of operational variables

A small increase in the pH near the pKa (pKa = 9.24 (Skoog, West, Holler, & Crouch, 2000)) at which a shift occurs from ammonium to ammonia will cause a significant decrease in the ammonia removal efficiency (Figure 4.9). The

removal efficiency of H_2S, being an acid (pKa = 7.02 (Skoog et al., 2000)), is influenced by the pH in the opposite way as for NH_3 (results not shown). Nevertheless, the maximum removal efficiency that could be reached, at pH = 14, was still only 1.4% compared to 0% in the reference case. This is related to the high Henry coefficient of H_2S (low solubility) and the low residence time applicable in air scrubbers for agricultural use.

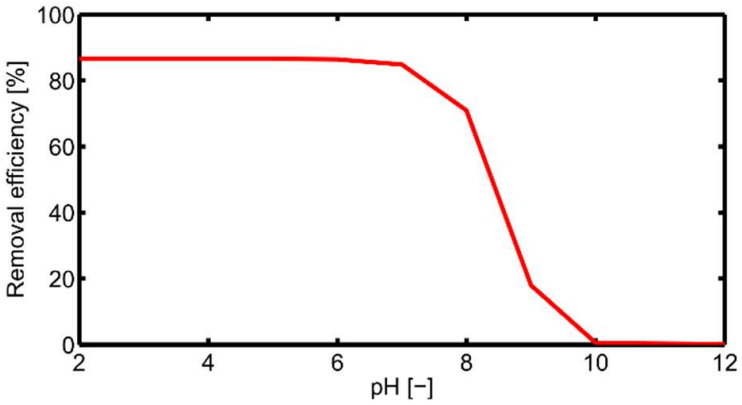

Figure 4.9. Simulated influence of the pH on the ammonia removal efficiency.

At increasing liquid flow rate, the mass transfer coefficient increased, leading to an increased removal efficiency (results not shown). However, the latter increase was very small (only 1.2% between 0.4 and 35 $m^3.(m^2.h)^{-1}$). Since only a very small improvement in removal efficiency is observed with increasing liquid flow rates, the flow rate can be kept as low as possible to reduce the pumping costs for recirculation, while keeping the packing material completely wet.

4.7 Conclusions

A mechanistic model for a countercurrent chemical air scrubber was set up based on incoming concentrations of pollutants, air temperature and relative humidity. Model evaluation through simulation showed the following:

- The model is able to predict the removal efficiency, the gas and liquid temperature profile, the relative humidity profile and the evaporation rate

at the same time. It thus provides a useful tool for further process optimization in terms of design and control.

- The choice of the correlations for both gas and liquid mass transfer coefficients significantly affects the simulation results; its selection requires specific attention.
- The model was validated for typical conditions prevailing in pig housing facilities in Flanders. The model was able to describe the experimental results with an average difference for outgoing ammonia concentration, air temperature and relative humidity of 1.2 ppm, 1.1°C. and 5.0%, respectively.
- The ammonia removal efficiency increases with decreasing pH of the washing liquid as well as with an increasing packing volume or specific surface area. A higher increase is achieved by increasing the packing height rather than its length or width, for the same contact surface area.
- The water consumption increases with increasing temperature and decreasing relative humidity of the incoming gas stream, while they hardly affect the removal efficiency.
- The ventilation rate has a significant influence on both the removal efficiency and the water consumption. Higher ventilation rates lead to a decreasing removal efficiency because of a decreased contact time rather than because of dilution of the incoming air.

Appendix 4A: Temperature dependencies

The temperature dependency of the density of water, the heat capacity of air and water and the enthalpy of vaporization of water were found very small, with less than 3% between values of 1°C and 40°C and were therefore not introduced in the model.

$$H = \frac{1}{k_H^{\circ} \cdot R \cdot (T_{\text{int}} + 273.15) \cdot \exp\left(\frac{\Delta_{sol}H}{R} \cdot \left(\frac{1}{(T_{\text{int}} + 273.15)} - \frac{1}{(T_{ref} + 273.15)}\right)\right)} \quad \text{G/W} \quad \textbf{Eq.S4.1}$$

$$K_{a,NH_4^+} = 10^{-\left(0.09018 + \frac{2729.82}{T_L + 273.15}\right)} \cdot 1000 \quad [1] \quad \textbf{Eq.S4.2}$$

$$p_{sv}^i = 133.29 \cdot \exp\left[18.30 - \frac{3816.44}{(T_G^i + 273.15) - 46.13}\right] \quad [2] \quad \textbf{Eq.S4.3}$$

$$\rho_G = \frac{(p_{atm} - p_v) \cdot M_{DA} + p_v \cdot M_{H_2O}}{R \cdot (T_G + 273.15) \cdot 1000 g.kg^{-1}} \quad \textbf{Eq.S4.4}$$

$$\mu_G = \frac{1.458 \cdot 10^{-6} \cdot \left(T_{\text{int}} + 273.15\right)^{1.5}}{\left(T_{\text{int}} + 273.15\right) + 110.4} \quad [3] \quad \textbf{Eq.S4.5}$$

$$\mu_L = 1602.4 \cdot \exp\left[-0.023 \cdot T_{\text{int}}\right] \cdot 10^{-6} \quad [4] \quad \textbf{Eq.S4.6}$$

$$\lambda_G = 0.02624 \cdot \left(\frac{T_{\text{int}}}{300}\right)^{0.8646} \quad [3] \quad \textbf{Eq.S4.7}$$

$$\lambda_L = 0.0017 \cdot T_{\text{int}} + 0.5637 \quad [4] \quad \textbf{Eq.S4.8}$$

$$D_{G,C} = \frac{10^{-7} \cdot (T_G + 273.15)^{1.75} \cdot \left(1/M_{NH_3} + 1/M_{DA}\right)^{1/2}}{P\left[\left(\sum_C \upsilon_i\right)^{1/3} + \left(\sum_{air} \upsilon_i\right)^{1/3}\right]^2} \quad [5] \quad \textbf{Eq.S4.9}$$

$$D_{L,C} = 7.4 \cdot 10^{-12} \left[\left(2.6 \cdot M_{H_2O}\right)^{1/2} \frac{\cdot (T_L + 273.15)}{\mu_{H_2O} \cdot 1000 \cdot {\upsilon_c}^{0.6}}\right] \quad [6] \quad \textbf{Eq.S4.10}$$

[1] (Emerson, Russo, Lund, & Thurston, 1975)
[2] (Fazaelipoor, 2010)
[3] (Dixon, 2007)
[4] (Lemmon, McLinden, & Friend, 2013);
Fit between 0 and 50°C, R^2 higher than 0.99
[5] (Fuller, Schelter, & Giddings, 1966)
[6] (Wilke & Chang, 1955)

Appendix 4B: Correlations for the mass transfer coefficient

Billet & Schultes (1999):

$$k_G = cp_G \cdot \frac{a^{0.5} \cdot D_G}{\sqrt{d_h(\varepsilon - h_L)}} \cdot \left(\frac{\rho_G \cdot u_G}{a \cdot \mu_G}\right)^{3/4} \cdot Sc^{1/3} \qquad \textbf{Eq.S4.11}$$

$$k_L = cp_L \cdot \left(\frac{\mu_L \cdot g}{\rho_L}\right)^{1/6} \cdot \left(\frac{D_L}{d_h}\right)^{0.5} \left(\frac{u_L}{a}\right)^{1/3} \qquad \textbf{Eq.S4.12}$$

Onda et al. (1968):

$$k_G = 5.23 \cdot a \cdot D_G \cdot \left(\frac{u_G \cdot \rho_G}{a \cdot \mu_G}\right)^{0.7} \cdot \left(\frac{\mu_G}{\rho_G \cdot D_G}\right)^{1/3} \cdot (a \cdot d_p)^{-2.0} \qquad \textbf{Eq.S4.13}$$

$$k_L = 0.0051 \cdot \left(\frac{\rho_L}{\mu_L \cdot g}\right)^{-1/3} \cdot \left(\frac{u_L \cdot \rho_L}{a \cdot \mu_L}\right)^{2/3} \cdot \left(\frac{\mu_L}{\rho_L \cdot D_L}\right)^{-0.5} \cdot (a \cdot d_p)^{0.4} \qquad \textbf{Eq.S4.14}$$

Shulman et al. (1955):

$$k_G = 1.95 \cdot u_G \cdot \left(\frac{d_p \cdot u_G \cdot \rho_G}{\mu_G(1-\varepsilon)}\right)^{-0.36} \cdot Sc^{-2./3} \qquad \textbf{Eq.S4.15}$$

$$k_L = 25.1 \cdot \frac{D_L}{d_p} \cdot \left(\frac{d_p \cdot u_L \cdot \rho_L}{\mu_L}\right)^{-0.36} \cdot Sc^{0.5} \qquad \textbf{Eq.S4.16}$$

Appendix 4C: Estimation of maximum volume change of the liquid phase over the scrubber

Consider the extreme situation of a warm and dry exhaust gas (35°C and relative humidity of 45%) which cools down to 5°C and increases in relative humidity to 99%. The water loss amounts then to 130 gH_2O per m^3 air (Eq.4.6, Eq.4.7 and Eq.S4.3). At a gas volume of 0.27 m^3 in each cell, this implies a maximal evaporation of 35 g or 35.10^{-6} m^3 water per cell. Considering a liquid volume of 0.27 m^3 in each cell, the maximum difference in liquid volume due to evaporation in a cell is 0.013% and for 20 cells in the packing 0.26%. The difference in volume between the bottom and upper cell thus may be neglected.

5

Mechanistic modelling of a biological air scrubber

5.1 Abstract

A mechanistic model for ammonia removal in a countercurrent biological air scrubber was set up. Besides mass balances of the components participating in the biological conversions, the water mass balance and the heat balance were considered as well. The effects of the pH and the concentration of the nitrogen components on the driving force for mass transfer were scrutinized. The model was compared against experimental data from a pig housing facility. Simulations were performed to assess the usefulness of pH control and to investigate the effect of inlet air conditions on the ammonia removal efficiency.

Van der Heyden, C., Solon, K., De Keukeleire, L., Demeyer, P. & Volcke, E.I.P. (in preparation). Model-based evaluation of ammonia removal in biological air scrubbers.

5.2 Introduction

Biofiltration by biological air scrubbers or so-called biotrickling filters has evolved to an off-shelf technique to treat exhaust air from industry but also from mechanically ventilated animal houses, as they can successfully be applied for large air streams with low concentrations (Cox & Deshusses, 1998). The contaminated air is passed through a packed bed where soluble and biodegradable gases such as ammonia or volatile compounds are absorbed and converted by bacteria which are part of the biofilm attached to the packing material. Ammonia is converted by nitrification, i.e. its biological conversion into nitrite and subsequently into nitrate. Despite the relative simple working principle of biotrickling filters, biofiltration is a complex process involving several physical, chemical and biological interactions.

In biological air scrubbers, the washing water which is recirculated over the packing needs to be regularly refreshed to ensure proper operation. The water discharge can be controlled automatically by measuring electrical conductivity (EC), as it is correlated to the total nitrogen concentration, TN, including ammoniacal, nitrite and nitrate nitrogen (Melse, Ploegaert et al., 2012, Chapter 6). When the predefined EC setpoint value is reached, a fixed small volume of water is discharged from the buffer tank, using a time-controlled valve, and replaced by fresh water. In this way the nitrogen content in the buffer tank only decreases slightly, keeping it more or less constant. Additionally, pH should remain in the optimal range for microbial growth, i.e. between 6.5 and 8 (Van Hulle et al., 2007). According to the Dutch and Flemish legislation, the pH must at least remain between 6.5 and 7.5 for proper operation (InfoMil, 2015; MB31/05/2011). However, biological air scrubbers installed at pig housing facilities in Flanders and the Netherlands most often do not have a pH control system, while in Germany, acid and/or base are dosed at biological air scrubbers when necessary (DLG, 2014).

Continuous short-term and long-term ammonia measurements at biological air scrubbers applied in pig housing facilities, showed a daily and seasonal pattern in the ammonia removal performance (Melse, Ploegaert, et al., 2012; Van der

Heyden, Brusselman, Volcke, & Demeyer, 2016). These changes could be attributed to a fluctuating ventilation rate, loading rate or air temperature. The ventilation rate in a pig housing facility determines the air flow rate through the air scrubber and is controlled based on the air temperature inside the housing facility (Van Gansbeke et al., 2009). Besides, an increased ventilation rate results in a lower ammonia inlet concentration due to dilution if the emission rate is constant. Nevertheless, it is also possible that with increasing flow, more ammonia is extracted from the building due to an increased ammonia emission by emitting surfaces and increased defecation and urination (Melse, Ploegaert, et al., 2012). Because of the interrelation between ventilation rate, air inlet temperature and loading rate, it is not fully clear which are the primary variables responsible for the variation in biotrickling filter performances.

The air flow through an air scrubber is inversely proportional to the empty bed residence time (EBRT) or contact time through the air scrubber and thus may negatively affect the removal efficiency. The air temperature is expected to have a negative influence on the removal efficiency as it influences the ammonia solubility, expressed as the Henry's law constant H, which decreases by 5% for a temperature rise of 1°C (Sander, 1999). Melse, Ploegaert et al. (2012) performed a long term measurement campaign and hypothesized that air temperature is the main influencing disturbance variable, rather than the changing ventilation rate and load. However, it is difficult to test the relative importance of these variables in practice as they are interrelated.

In this chapter, the behaviour of a biological air scrubber was described through a mechanistic model. Ammonia removal, temperature and evaporation dynamics were described through mass and energy balances. Particular attention was paid to the effects of pH and the concentration of nitrogen components in the washing water on the driving force for mass transfer. Simulations were carried out, comparing open-loop operation with pH control. The latter was used to study the effect of influent characteristics: air temperature, ventilation rate and ammonia load.

5.3 Materials and methods

5.3.1 Operating principle of a biological air scrubber

The physicochemical and biological conversions associated with a biological air scrubber for ammonia removal are schematically represented in Figure 5.1. Ammonia is absorbed from the gas phase into the liquid phase, where a chemical equilibrium with ammonium is established, which depends on the pH. The ammonia concentration is reduced by nitrification, i.e. biological conversion to nitrite and subsequently to nitrate.

The driving force (*DF)* for ammonia mass transfer from the gas to the liquid phase, is determined by the ammonia concentration in the gas phase, which is in equilibrium with its concentration in the liquid phase $C_{L,NH3}$* and the ammonia concentration in the liquid phase $C_{L,NH3}$:

$$DF = C^*_{L,NH_3} - C_{L,NH_3} = \frac{C_{G,NH_3}}{H} - C_{L,NH_3} = \frac{C_{G,NH_3}}{H} - C_{L,TAN}\left(1 - \frac{C_{H^+}}{C_{H^+} + Ka_{NH_3}}\right) \quad \textbf{Eq.5.1}$$

The effect of pH and of the total ammoniacal nitrogen concentration in the washing water ($C_{L,TAN}$) on the driving force for mass transfer was studied to gain insight in the relative importance of both variables.

The ammonia absorption and nitrification processes are both pH dependent and effect the pH at the same time. The absorption of 1 mole of ammonia involves the consumption of 1 mole of protons (indicated by C, Figure 5.1), resulting in a pH increase. Nitrification results in the production of 2 moles of protons per mole ammonia converted (indicated by B, Figure 5.1), thus decreasing pH. Nitrification stops when the pH decreases below a value where the substrate (ammonia) is no longer available (indirect effect) and that inhibits nitrification (direct effect) (Van Hulle et al., 2007).

It is often stated that the role of nitrification in a biological air scrubber for ammonia removal is to increase the driving force for mass transfer from the gas phase to the liquid phase by reducing the concentration of total ammoniacal ammonium ($C_{L,TAN}$; indicated by A in Figure 5.1). Additionally, nitrification also results in a pH decrease, shifting the chemical equilibrium to a relatively

lower concentration of free ammonia ($C_{L,NH3}$; indicated by B in Figure 5.1), additionally increasing the driving force for ammonia transfer from the gas phase to the liquid phase.

Therefore, the effect of nitrification through the reduction of total ammonium and through the decrease of pH were considered separately, to investigate their respective impact on the driving force. For this purpose, a batch of washing water was considered, containing a total nitrogen content of 3.2 $gN.L^{-1}$ (i.e., the maximum value allowed in Flanders, (MB31/05/2011), distributed as 50% TAN and 50% nitrate. An ammonia gas concentration of 20 ppm was assumed; the corresponding equilibrium liquid concentration was considered in the calculation of the driving force for ammonia mass transfer ($C_L^*{}_{,NH3} = C_{G,NH3}/H$). A temperature of 20°C was assumed. The change in pH was iteratively calculated through the charge balance method (Appendix 5.A.2, Eq.S5.1), considering also the bicarbonate/carbon dioxide equilibrium, assuming that the washing water is saturated with CO_2 (508 $gC.m^{-3}$).

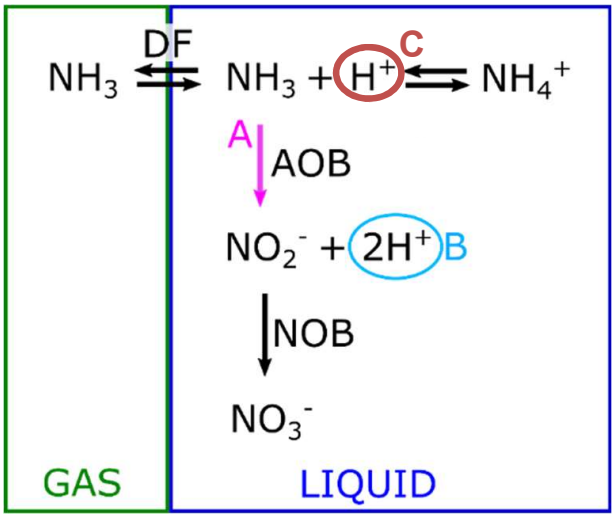

Figure 5.1. Physicochemical and biological conversions in a biotrickling filter for ammonia removal. Nitrification results in a reduction of the ammonia concentration (A) as well as in a pH decrease (B). Absorption of ammonia from the gas into the liquid phase results in a pH increase (C). DF: driving force

5.3.2 Biological air scrubber model

A mechanistic model for a biological countercurrent air scrubber was set up and implemented in Matlab-Simulink®. The physical-chemical part of the model was based on the model of a chemical air scrubber by Van der Heyden et al. (2016). The packing material was divided into a number (nY = 10) of horizontal cells, complying with the required accuracy and still keeping a reasonable calculation time. A fixed-step discrete solver was used with a time step chosen smaller than the refreshment rate of each cell. Each cell consisted of an air and liquid phase, each of which was modelled as an ideally mixed continuous stirred-tank reactors (CSTR), which were separated by a stagnant boundary layer at the interface (two film theory) (Figure 4.1).

Both mass transfer and heat transfer were considered, allowing to simulate the removal efficiency, temperature and evaporation in the air scrubber. The energy and water balances, including the temperature dependencies of the used parameters, were similar as in the chemical air scrubber model (Van der Heyden, Vanthillo, et al., 2016). The individual gas and liquid phase mass balances for a component with gas phase concentration C_G and liquid concentration C_L (additional index to specify component is omitted for simplicity) in cell i were implemented as

$$\frac{dC_G^i}{dt} = \frac{Q_G}{V_{G,cell}} \cdot \left(C_G^{i+1} - C_G^i\right) - K_L^i\left[T_G^i, T_L^i\right] \cdot a \cdot \left(\frac{C_G^i}{H^i\left[T_G^i, T_L^i\right]} - C_L^i\right) \qquad \textbf{Eq.5.2}$$

$$\frac{dC_L^i}{dt} = \frac{Q_L}{V_{L,cell}} \cdot \left(C_L^{i-1} - C_L^i\right) + K_L^i\left[T_G^i, T_L^i\right] \cdot a \cdot \left(\frac{C_G^i}{H^i\left[T_G^i, T_L^i\right]} - C_L^i\right) - r_b \qquad \textbf{Eq.5.3}$$

Respectively. Eq.5.2 expresses that the accumulation or depletion of a component in the gas phase in cell i is due to the influx from the underlying cell with concentration C_G^{i+1} and the outflow to the overlying cell with concentration C_G^{i}, as well as transfer from this pollutant from the gas phase to the liquid phase or vice versa. K_L is the total mass transfer coefficient, considering the resistances at the liquid and gas interface, calculated using the correlation of Billet & Schultes (1999). Eq.5.3 expresses the liquid phase concentration dynamics in an analogous way.

The individual biomass mass balance in cell i was implemented as

$$\frac{dX^i}{dt} = \frac{\tau \cdot Q_L}{V_{L,cell}} \cdot \left(X^{i+1} - X^i\right) + r_b \qquad \textbf{Eq.5.4}$$

with τ the biomass retention factor. r_b denotes the conversion rate of component b, which is calculated from the process rates and the stoichiometric matrix Table S5.1 (Appendix 5.A.1), through Eq.5.5.

$$r_b = \sum_j A_{bj} R_j \qquad \textbf{Eq.5.5}$$

The components considered in the gas phase were ammonia (NH_3), oxygen (O_2) and carbon dioxide (CO_2). The components considered in the liquid phase were ammonia (NH_3) and ammonium (NH_4^+), lumped into total ammonia nitrogen (TAN), nitrite (NO_2^-) and nitrous acid (HNO_2), lumped into total nitrite nitrogen (TNO_2), nitrate (NO_3^-), carbon dioxide (CO_2), bicarbonate (HCO_3^-) and carbonate (CO_3^{-2}), lumped into total carbon (TIC), as well as oxygen (O_2). Two types of bacteria were considered in the biofilm and in the liquid phase, namely ammonia oxidizing bacteria (X_{AOB}) and nitrite oxidizing bacteria (X_{NOB}). Heterotrophic growth was not considered.

In a biotrickling filter, the bacteria carrying out the biological conversions are mostly attached to the packing material. The retention of bacteria on the biotrickling filter packing was modelled by including a retention factor in the biomass mass balance, thus neglecting spatial biofilm gradients. This approach was justified as the purpose of the model is to study the overall reactor performance. The applicability of this type of zero-dimensional models for the description of a moving bed biofilm reactor has been demonstrated before (Plattes, Henry, & Schosseler, 2008).

The process rate regarding nitritation and nitratation is expressed by a Monod-equation taking into account the affinity for O_2 and NH_3 and NO_2. Inhibition by free ammonia (FA) and free nitrous acid (FNA) was assumed to be negligible in this study as nitrite did not accumulate in the study of Melse, Ploegaert et al. (2012). The biomass decay rate was described as directly

proportional with the biomass concentration. The stoichiometric and kinetic parameters related to AOB and NOB growth are summarized in Table S5.2 (Appendix 5.A.1). The effect of temperature on the maximum specific growth rate and decay rate is taken into account, considering the Arrhenius equation (Hao, Heijnen, & van Loosdrecht, 2002):

$$r_T = r_{293} \cdot \exp\left[\frac{-E_{act} \cdot (293 - T)}{R \cdot 293 \cdot T}\right] \qquad \textbf{Eq.5.6}$$

Absorption of ammonia and biological conversion reactions that involve proton consumption or production affect the pH of the medium. Vice versa, pH influences the biological conversion rates both through a direct effect on the maximum growth rates (Van Hulle et al., 2007) and through the chemical equilibria of the species involved. pH was therefore added in the model as a state variable and calculated by means of the charge balance method (Hellinga et al., 1999; Volcke, 2006), as detailed in Appendix 5.A.2.

The bacteria are indirectly dependent on the pH due to the equilibrium between ammonia and ammonium. A direct pH-dependency of the maximum specific growth rate was added, according to Van Hulle et al. (2007):

$$\mu = \mu_{\max} \cdot \frac{K_{pH}}{K_{pH} - 1 + 10^{|pH_{opt} - pH|}} \qquad \textbf{Eq.5.7}$$

The optimum pH range for bacterial growth was characterized by a pH optimum of 7.23 (pH_{opt}, corresponding with maximum bacterial growth) and by a parameter K_{pH} = 8.21. This description resulted in a bacterial growth rate which was at least 50% of the maximum value in the pH range from 6 to 8.5 (see Appendix 5.A.3).

Eq.5.2 and Eq.5.3 are valid in all cells except the top and bottom compartments. The washing water sprayed on top of the packing, originates from the buffer tank and thus has the same composition (Eq.5.8). The gas phase concentration at the bottom cell is equal to the incoming concentration (Eq.5.9). In the top and bottom cell, no mass transfer between gas and liquid is considered.

$$C_L^1 = C_L^{BT} \quad \text{Eq.5.8}$$

$$C_G^{nY+2} = C_G^{in} \quad \text{Eq.5.9}$$

In biological air scrubbers, the discharge strategy to keep a constant EC value or total nitrogen content, is frequently used in practice. In the model, this discharge strategy is implemented by setting a maximum allowed total nitrogen concentration in the buffer tank ($C_{BT,TN}^{max}$) and a minimal total nitrogen concentration ($C_{BT,TN}^{min}$), which is close to the maximum allowed concentration. When $C_{BT,TN}^{max}$ is reached in the buffer tank, the concentration in the buffer tank is instantaneously decreased to $C_{BT,TN}^{min}$ at the next model iteration. The total nitrogen concentration will then increase again to $C_{BT,TN}^{max}$ for the next discharge. This discharge strategy implies that the total amount of absorbed ammonia from the gas into the liquid phase, is discharged to keep the total nitrogen at the desired setpoint. The discharge rate can thus be calculated taking into account the absorbed ammonia concentration and the maximum nitrogen concentration in the washing water, according to following equation:

$$Q_D = \frac{C_{G,N}^{in} \cdot Q_{vent} \cdot \eta / 100}{C_{L,N}^{\max}} \quad \text{Eq.5.10}$$

5.3.3 System under study and simulation set-up

A reference case was defined based on the biotrickling filter studied by Melse, Ploegaert, et al. (2012). It concerns a countercurrent biotrickling filter to treat the exhaust of a pig housing facility of 600 sows, with a maximum of 40 000 $m^3.h^{-1}$ ventilation air. The inlet conditions were set according to average values, measured by Melse, Ploegaert, et al. (2012) during a 141 days measurement period (Table 5.1). Discharge was automatically controlled using an electrical conductivity (EC) setpoint around 16 $mS.cm^{-1}$ to limit the total nitrogen content in the washing water, which varied between 1775 and 4817 $gN.m^{-3}$. The discharge rate was on average 0.33 $m^3.d^1$.

In the model, the maximum allowed total nitrogen content in the washing water

was set at 3200 gN.m^{-3} ($C_{BT,TN}^{max}$), which is the prescribed setpoint in Flanders (MB31/05/2011) and an acceptable average of the maximum total nitrogen concentrations as found by Melse, Ploegaert et al. (2012). The minimal total nitrogen concentration in the buffer tank after discharge ($C_{BT,TN}^{min}$) was set to 3150 gN.m^{-3}. The discharge rate to keep the total nitrogen concentration below this setpoint amounts to 0.80 m^3.d^{-1} (Eq. 5.10).

The washing water pH was not controlled in the study of Melse, Ploegaert et al. (2012). The reference case behaviour was simulated therefore with the model including pH dynamics. The initial pH was set to 6.6, which is the average pH value measured by Melse, Ploegaert et al. (2012).

Table 5. 1. Reference case - design and input parameters (Melse, Ploegaert et al., 2012)

Symbol	Description	Value	Unit
H_P	Packing height	1.1	m
A_P	Packing cross sectional area	3.8	m^2
A_{spec}	Packing specific surface area	100	m^2.m^{-3}
V_{BT}	Buffer tank volume	20	m^3
$T_{G,in}$	Incoming air temperature	20	°C
T_{Lin}	Incoming liquid temperature (assumed)	13	°C
$C_{G,NH3,in}$	Incoming ammonia concentration	14	ppm
Q_G	Gas flow rate	13900	m^3.h^{-1}
Q_L	Liquid flow rate	15	m^3.h^{-1}
$L_{NH3,in}$	Incoming ammonia load	396	gN.h^{-1}

Table 5. 2. Initial conditions

Variable	Description	Value	Unit
T_G	Initial temperature gas phase	20	°C
T_L	Initial temperature liquid phase	13	°C
T_{BT}	Initial temperature buffer tank	13	°C
RH	Initial relative humidity gas phase	65	%
$C_{G,NH3}$	Initial NH_3 gas concentration	0	gN.m^{-3}
$C_{L,TN}$	Initial TN concentration liquid phase	0	gN.m^{-3}
$C_{BT,TN}$	Initial TN concentration buffer tank	0	gN.m^{-3}
$C_{L,AOB}$	Initial AOB concentration liquid phase	5000	gCOD.m^{-3}
$C_{L,NOB}$	Initial NOB concentration liquid phase	5000	gCOD.m^{-3}

The scenario without pH control (open-loop behaviour) was subsequently compared to a system with pH control at a constant setpoint of 6.6. This model was then applied to study the influence of the air inlet characteristics on the ammonia removal efficiency, by independently varying the air temperature, ventilation rate and ammonia loading rate.

5.4 Results and discussion

5.4.1 Effect of washing water pH and nitrogen component concentrations on ammonia mass transfer

The total nitrogen content in the washing water is determined by the discharge setpoint and corresponds with a certain total ammoniacal nitrogen (TAN) concentration. The TAN concentration and pH at their turn determine the amount of free ammonia in the water phase ($C_{L,NH3}$, Eq.5.1). In order to have a driving force for ammonia mass transfer from the gas phase to the liquid phase, the ammonia concentration in the liquid phase ($C_{L,NH3}$) needs to be lower than the equilibrium liquid ammonia concentration corresponding with the ammonia concentration in the gas phase, which is calculated using the Henry coefficient ($C_L^*{}_{,NH3} = C_{G,NH3}/H$). Figure 5.2 displays the free ammonia concentration in the liquid phase ($C_{L,NH3}$) as a function of pH and various total ammoniacal nitrogen concentrations ($C_{L,TAN}$) and confronts it with the equilibrium concentration for various ammonia gas phase concentrations ($C_L^*{}_{,NH3}$). The driving force is only positive in case $C_L^*{}_{,NH3}$ is higher than $C_{L,NH3}$.

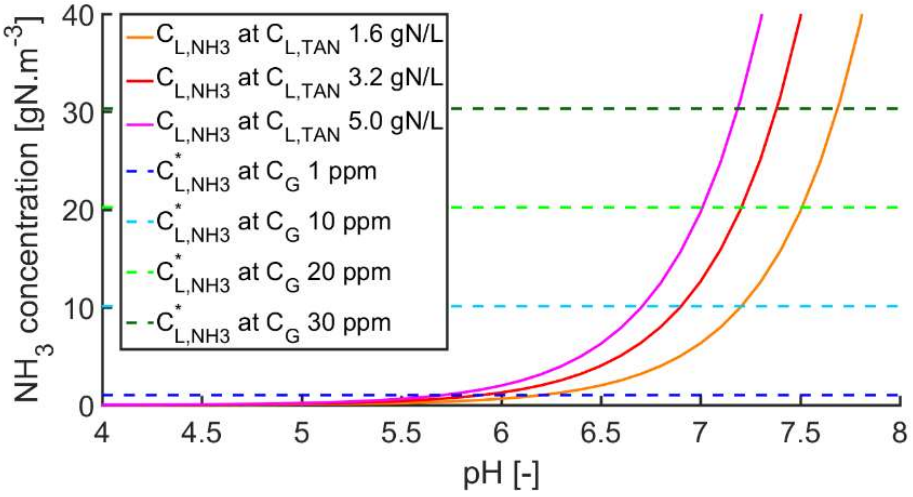

Figure 5.2. Free ammonia concentrations in the washing water as a function of pH and various total ammoniacal nitrogen concentrations ($C_{L,NH3}$; solid lines); ammonia liquid concentrations in equilibrium with various gas concentrations, based on Henry's law ($C_L^*{}_{,NH3}$; dashed lines)

For example at a pH of 7, ammonia at a gas concentration of 10 ppm, can be removed if the TAN concentration is 1.6 $gN.L^{-1}$ but this is no longer the case

at 3.2gN.L^{-1}. This shows that it is important to keep the total nitrogen setpoint, and in this way the total ammoniacal nitrogen content in the washing water, low enough. Lower pH values of the washing water allow higher total nitrogen concentrations without compromising the driving force for ammonia transfer.

Besides, the higher operating pH in a biological air scrubber compared to a chemical air scrubber has a strong effect on the driving force for ammonia from the gas phase to the liquid phase. At a pH of 4 or lower, as applied in a chemical air scrubber, all total ammoniacal nitrogen is in the form of ammonium. Therefore, the total allowed nitrogen content in the washing water may be significantly higher than in biological air scrubbers (58 gN.L^{-1} compared to 3.2 gN.L^{-1}; (MB31/05/2011), reducing the discharge rate significantly (Eq.5.10). In a biological air scrubber, such high total nitrogen concentrations cannot be allowed. As the pH lies only around 7, a slight increase can immediately result in a driving force below zero.

The total effect of nitrification concerns a decreased total ammoniacal nitrogen concentration and a decreased pH, which both have a positive effect on the driving force for ammonia transfer (solid lines in Figure 5.3). The driving force for ammonia transfer (*DF*) increases with increasing ammonia conversion, until it reaches the maximum value equalling $C_L^*{}_{,NH3}$. At this point, no free ammonia (FA) is present in the liquid as the pH is low (pH < 4) and the ammonia/ammonium equilibrium shifts completely to ammonium ($pK_{a,NH3}$ = 9.4 at 20°C).

The effect of nitrification on the driving force for ammonia mass transfer (DF) was also studied for a constant pH, considering only the decreased total ammonium concentration (dashed lines in Figure 5.3). In this case, the driving force increases linearly with an increasing ammonium conversion, at a small slope. Comparing the two scenarios, it is clear that the effect of nitrification on the driving force for ammonia mass transfer is mainly due the associated pH decrease rather than due to the reduction of the total ammonium concentration in the liquid.

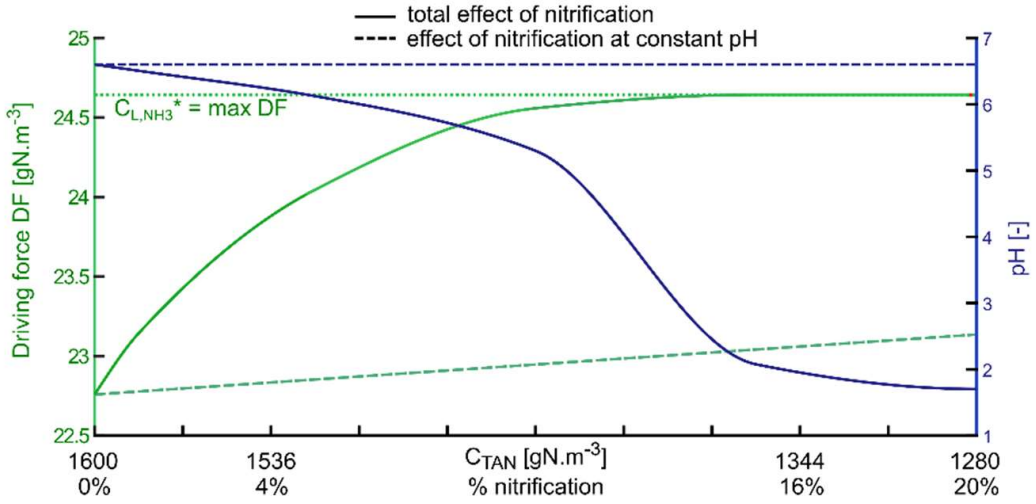

Figure 5.3. Effect of nitrification on the driving force for ammonia mass transfer (DF), comparing the total effect of nitrification (A + B, Figure 5.1) with the effect of nitrification at constant pH (A, Figure 5.1) . DF and pH are displayed as a function of the percentage of the total ammoniacal nitrogen concentration which is nitrified.

The effect of pH will even be more pronounced when the initial pH of the liquid batch is higher because the initial driving force is smaller in this case. If 1% is nitrified at an initial pH of 7.5, this induces a decrease of pH by only 0.2, but this doubles the initial driving force.

In practice, the pH will not drop below 4 as nitrification has a pH optimum and is inhibited at low pH (Van Hulle et al., 2007). Additionally, these results only represent a batch test. During normal (continuous) operation of an air scrubber, absorption of ammonia from the gas to the liquid phase will increase the pH of the washing water.

5.4.2 Reference case without pH control

The scenario without pH control resulted in a steady state ammonia removal efficiency of 92%. The total ammoniacal nitrogen, total nitrite and nitrate concentrations were 1.75, 0.001 and 1.45 gN.L^{-1}, respectively, corresponding with a total nitrogen concentration of 3.2 gN.L^{-1} (Figure 5.4). Only half of the total ammonia concentration in the water was converted, as 54.5% of the total nitrogen is still total ammoniacal nitrogen. The observed incomplete ammonia conversion was attributed to the effect of pH. As nitrification produces two moles of protons, while absorption only consumes one mole of protons, the pH decreases once bacteria have converted half of the absorbed ammonia, limiting

further ammonia conversion. An equilibrium between ammonia absorption and nitrification is reached at a steady state pH of 6.2.

The nitrate concentration is high (44.7% of the total nitrogen concentration) while the nitrite concentration in the washing water is very low (0.8% of the total nitrogen concentration; Figure 5.4), indicating that almost all nitrite, which is formed by the AOB, is immediately converted by NOB. The AOB concentration is higher than the NOB concentration, as AOB have a higher specific yield than NOB.

The gas temperature inside the air scrubber decreased from 20°C at the inlet to 15°C at the outlet, thereby heating up the washing water from 13°C to 15°C. The relative humidity increased from 65% to almost 100%. These temperature and relative humidity profiles are similar to the ones for the chemical air scrubber model (Van der Heyden, Vanthillo, et al., 2016).

The simulation results agree well with the measured data of Melse, Ploegaert et al. (2012) (Figure 5.4 and Table S5.3 in Appendix 5B). The slightly higher simulated ammonia removal, compared to the average measured value over the entire measurement period (efficiency 92% simulated versus 82% measured), could be attributed to slightly higher average total nitrogen content (Figure 5.4) and to the dynamics in the operation of the air scrubber of Melse, Ploegaert et al. (2012). The distribution of the nitrogen components in the water was similar, with only half of the total nitrogen converted to nitrate. Incomplete nitrification was also observed during other measurement campaigns at pig housing facilities (Melse & Mosquera, 2014; Mosquera et al., 2011; Ottosen et al., 2011; Van der Heyden, Brusselman, et al., 2016). The simulated results thus agree well with the experimental data.

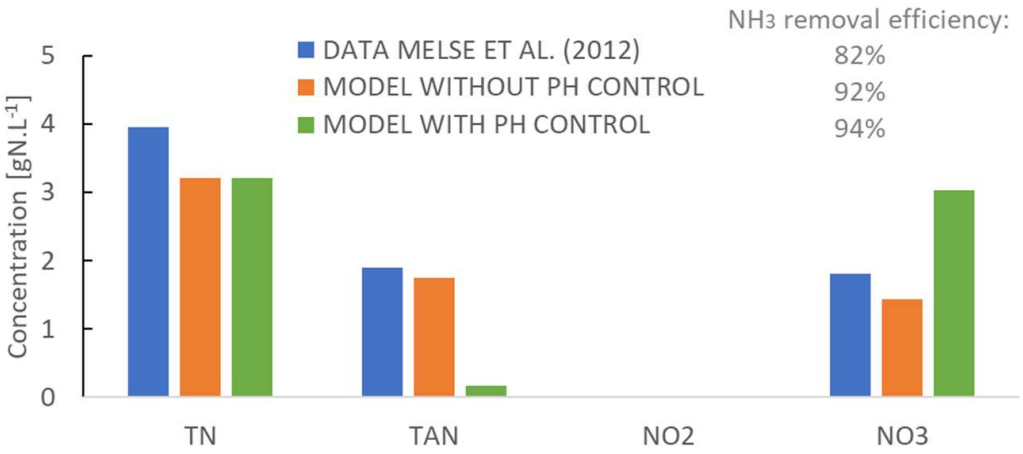

Figure 5.4. Comparison of the experimental data of Melse, Ploegaert et al. (2012) with the simulation results of model without pH control and the model with regulated pH, for the nitrogen components in the washing water: total nitrogen (TN), total ammoniacal nitrogen (TAN), nitrite (NO_2) and nitrate (NO_3)

5.4.3 Effect of pH control at a constant value

When the pH is controlled at a constant value of 6.6, almost all (99%) absorbed ammonia is converted to nitrate (Figure 5.4), instead of only 44.7% in case pH is not controlled. Base dosing, i.e. NaOH, avoids the pH decrease caused by and at its turn hampering nitrification, resulting in full conversion of ammonia to nitrate. Due to the dosing of a base, the pH can be kept constant, thereby allowing that nitrification exceeds the ammonia absorption. Besides, more AOB and NOB biomass is formed compared to the situation without pH control (Table S5.3 in Appendix 5B). Additionally, the driving force for ammonia mass transfer from the gas phase to the liquid phase is positively affected by the lower liquid phase ammonia concentration in case of pH control compared to the situation without pH control. However, only a slight increase in the ammonia removal efficiency was observed (94% with pH control at 6.6 compared to 92% without pH control).

Complete conversion of ammonia into nitrate in a biotrickling filter with pH control was demonstrated by Blázquez et al. (2017) at lab-scale, treating gas with an ammonia concentration of 100 ppmv, reaching a removal efficiency up to 99% and keeping the pH constant at 7 through on/off control based on the addition of NaOH (0.5M) or HCl (0.5M).

Increasing the pH setpoint in the range 5.8-8 only slightly increased the ammonia removal efficiency (from 91.6% to 94.2%, Figure 5.5). However, the distribution of nitrogen components in the washing water was strongly affected. For pH values of 7 or higher, about all nitrogen is converted to nitrate, but when the pH was kept at 5.8, all nitrogen in the washing water remained total ammoniacal nitrogen (Figure 5.5). It is clear that nitrification was limited or even completely inhibited outside the optimal pH range for bacterial growth. However, the ammonia removal efficiency remained high (92% at pH 5.8) even in the absence of nitrification, as all absorbed ammonia is immediately transformed to ammonium. In this case, the biological air scrubber functions as a chemical air scrubber. é

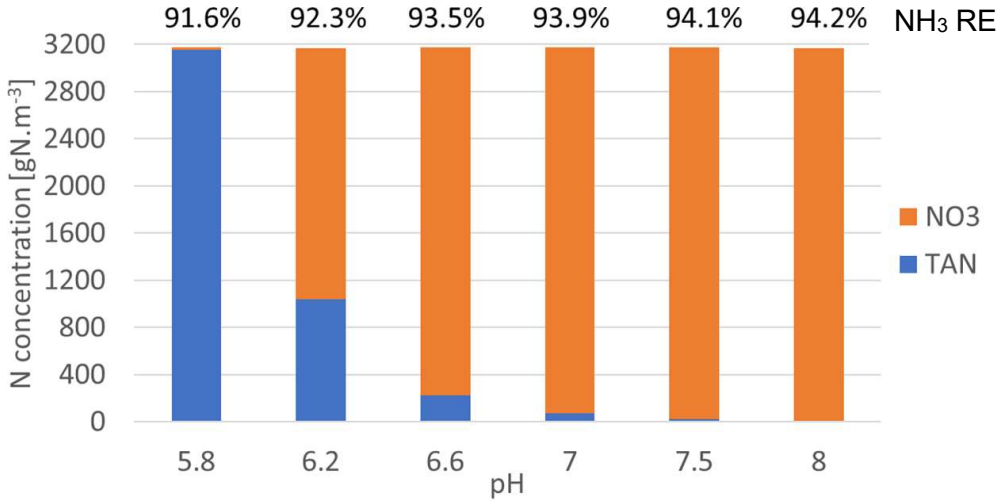

Figure 5.5. Simulated effect of pH setpoint on the ammonia removal efficiency and the concentrations of total ammoniacal nitrogen (TAN) and nitrate (NO_3) in the washing water. The total nitrite concentration (TNO_2) was zero in all cases.

The question arises to which extent the operation of a biological air scrubber with constant pH is different from a chemical air scrubber and if it is worth applying in practice. In a biological air scrubber the pH is controlled at neutral values compared to a pH of 4 or lower in a chemical air scrubber. The absorption and complete oxidation of one ammonia molecule results in a net production of one proton, which must be neutralized with one mole of base equivalent to keep a constant pH (Appendix 5C). Overall, base equivalents need to be added in an equimolar amounts compared to the ammonia absorbed. The amount of base equivalents that need to be dosed in a biological air scrubber with pH control and complete nitrification is thus equal to the amount

of acid equivalents that need to be dosed in a chemical air scrubber with pH control. It is clear that base addition for pH control in biological air scrubbers would increase the operational costs drastically, while only a slight increase in the ammonia removal efficiency is obtained compared to a biological air scrubber without pH control.

In sum, controlling the pH of a biological air scrubber at a constant value around 7 and stimulating full nitrification towards nitrate, seems not a valuable option to implement in practice in pig housing facilities. Nevertheless, it could be considered to install a pH control system to add acid in case the pH exceeds an upper threshold (pH above 7.5) to avoid a direct negative impact on the ammonia removal efficiency (Figure 5.2).

5.4.4 Effect of air inlet conditions on ammonia removal efficiency

The influence of the inlet air temperature, ventilation rate and ammonia load on the ammonia removal efficiency is summarized in Figure 5.6. The effect of an increased air temperature on the ammonia removal efficiency is negligible. At increased air temperatures, ammonia is less soluble (higher H), reducing the driving force to the liquid phase. The acidity constant increases (higher $K_{a,NH3}$), resulting in a higher concentration of ammonia compared to ammonium. This results in a decreased driving force as well as more ammonia availability for the biomass. Besides, the specific growth rate and decay rate of the biomass increase with increasing temperature (higher μ_{max} and b). Additionally, the mass transfer coefficient will increase (higher K_L), meaning that more ammonia will be transferred from the gas phase to the liquid phase and this has a positive effect on the removal efficiency. As a result of all these different effects of temperature, working in different ways, the overall effect on the ammonia removal efficiency is less than 1% over the considered temperature range.

At a higher ventilation rate or air flow rate through the air scrubber (Q in Figure 5.6), the incoming ammonia load increases but the EBRT (empty bed residence time = contact time) decreases. The net effect of both is a decreased ammonia removal efficiency.

When increasing the inlet ammonia load (L in Figure 5.6) while keeping a constant ventilation rate (i.e. by increasing the ammonia concentration), the outlet ammonia concentration increases but the removal efficiency remains constant.

The effects of air temperature, ventilation rate and load on the ammonia removal efficiency are similar to those for a chemical air scrubber (Van der Heyden, Vanthillo, et al., 2016). Overall, considering these simulation results, it is rather the ventilation rate than the inlet air temperature or the ammonia load that influences the ammonia removal efficiency in biological air scrubbers, which is different from the hypothesis of Melse, Ploegaert et al (2012). Dedicated experiments to confirm these findings could be performed at lab-scale or pilot-scale.

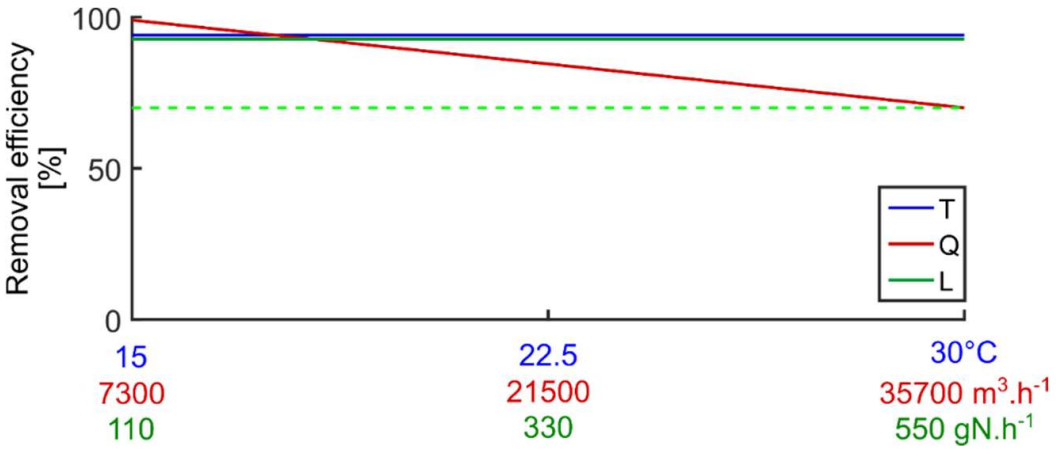

Figure 5.6. Influence of the inlet air temperature (T, °C), ventilation rate (Q, $m^3.h^{-1}$) and ammonia load (L, $gN.h^{-1}$) on the ammonia removal efficiency.

5.5 Conclusions

- A mechanistic model for a biotrickling filter for ammonia removal was set up, taking into account temperature, relative humidity and pH dynamics. The simulation results were in agreement with experimental data from a conventional pig housing facility.
- The positive impact of nitrification on the driving force for mass ammonia transfer is due to the associated pH decrease rather than by

the reduction of the total ammoniacal nitrogen content as such. The total nitrogen concentration in the washing water needs to be kept sufficiently low to sustain the driving force for ammonia transfer to the liquid phase.

- The application of pH control affects the nitrogen component distribution in the washing water rather than the ammonia removal efficiency. Without pH control, only half of the ammonia that is absorbed in the washing water can be oxidized by the biomass. When the pH is controlled at a constant value which is sufficiently low to allow ammonia mass transfer to the liquid phase but still high enough for bacterial growth, almost complete ammonium conversion can be achieved. However, the ammonia removal efficiency is hardly affected by pH control, while operational costs for base dosing are substantial.
- Simulation results clearly demonstrated that variations in the ammonia removal efficiency are mainly caused by a changing ventilation rate rather than by fluctuations in air temperature or ammonia load.

Appendix 5A: Biological air scrubber model

5.A.1. Stoichiometric matrix and parameter values

Table S5.1 Stoichiometric matrix.

A_{ij} **i component b →** **j process ↓**	$C_{L,NH3}$ [g N.m^{-3}]	$C_{L,HNO2}$ [g N.m^{-3}]	$C_{L,NO3}$ [g N.m^{-3}]	$C_{L,O2}$ [g O_2.m^{-3}]	X_{AOBi} [g COD.m^{-3}]	X_{NOB} [g COD.m^{-3}]	process rate ρ_j
1. ammonia oxidation - growth AOB	$-1/Y_{AOBi} - i_{NXB}$	$1/Y_{AOBi}$		$1-3.43/Y_{AOBi}$	1		$\rho_{G,AOB}$
2. nitrite oxidation – growth NOB	$-i_{NXB}$	$-1/Y_{NOB}$	$1/Y_{NOB}$	$1-1.14/Y_{NOB}$		1	$\rho_{D,AOB}$
3. decay.AOB	i_{NXB}				-1		$\rho_{G,NOB}$
4. decay NOB	i_{NXB}	$-(1-f_{XI})/1.71$				-1	$\rho_{D,NOB}$

$$R_{j,NH_3} = \mu_{\max}^{AOB} \cdot \frac{S_{b,O_2}}{K_{S,O_2}^{AOB} + S_{b,O_2}} \cdot \frac{S_{b,NH_3}}{K_{S,NH_3}^{AOB} + S_{b,NH_3}} \cdot X_{AOB}$$

$$R_{j,NO_2^-} = \mu_{\max}^{NOB} \cdot \frac{S_{b,O_2}}{K_{S,O_2}^{NOB} + S_{b,O_2}} \cdot \frac{S_{b,NO_2^-}}{K_{S,NO_2^-}^{NOB} + S_{b,NO_2^-}} \cdot X_{NOB}$$

$$R_{j,dAOB} = b^{AOB} \cdot X_{AOB}$$

$$R_{j,dNOB} = b^{NOB} \cdot X_{NOB}$$

Table S5.2. Stoichiometric and kinetic parameter values

Symbol	Description	Value (20°C)	Unit	Ref.
i_{NXB}	N-content of AOB and NOB	0.086	gN.gCOD^{-1}	[1]
$E_{act}^{nitritation}$	Activation energy of nitritation	65	kJ.mol^{-1}	[2]
$E_{act}^{nitratation}$	Activation energy of nitratation	45	kJ.mol^{-1}	[2]
AOB				
Y^{AOB}	Yield coefficient	0.18	gCOD.gN-NH_3^{-1}	[1]
$\mu_{\max}^{AOB}$	Maximum specific growth rate	0.8	d^{-1}	[2]
b^{AOB}	Specific decay rate	0.11	d^{-1}	[2]
K_{S,O_2}^{AOB}	Affinity constant of O_2	0.74	gO_2.m^{-3}	[3]
K_{S,NH_4}^{AOB}	Affinity constant for NH_3	0.25	gN-NH_3.m^{-3}	[4]
NOB				
Y^{NOB}	Yield coefficient	0.08	gCOD.gN-NO_2^{-1}	[1]
$\mu_{\max}^{NOB}$	Maximum specific growth rate	0.79	d^{-1}	[2]
b^{NOB}	Specific decay rate	0.11	d^{-1}	[2]
K_{S,O_2}^{NOB}	Affinity constant of O_2	1.75	gO_2.m^{-3}	[3]
K_{S,NO_2}^{NOB}	Affinity constant for NO_2^-	1.6	gN-NO_2.m^{-3}	[4]

[1] (Jubany, Baeza, Carrera, & Lafuente, 2007)
[2] (Hao et al., 2002)
[3] (Guisasola et al., 2006)
[4] (Henze et al., 1999)

5.A.2. Dynamic pH modelling

The pH profile along the air scrubber is obtained from the pH in each cell, which is calculated from the charge balance in that cell:

$$\Delta_{ch} = C_{H^+} - C_{OH^-} + C_{NH_4^+} - C_{NO_2^-} - C_{NO_3^-} - C_{HCO_3^-} - 2 \cdot C_{CO_3^{2-}} - C_{H_2PO_4^-} - 2 \cdot C_{HPO_4^{2-}} - 3 \cdot C_{PO_4^{3-}} + C_{Z^+} \qquad \textbf{Eq.S5.1}$$

The charged nitrogen ions ammonium, nitrite and nitrate are considered. The buffer capacity of the water was taken into account, considering also the bicarbonate/carbon dioxide equilibrium. Phosphate components were not considered. Z+ is an artificial component, which represents the amount of net positive charges, that are not influenced by the establishment of an equilibrium pH and that are not involved in any biological conversion reaction. It is calculated from the known initial pH and concentrations. The charge balance is implemented as in Volcke (2006) and is calculated by iteration, using the Newton-Raphson method.

5.A.3. Direct pH-dependency of the maximum specific growth rate

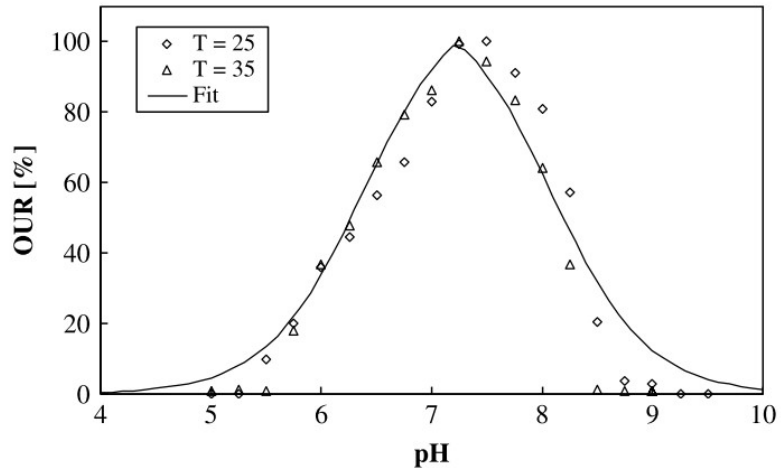

Figure S5.1. Influence of pH on oxygen uptake rate (OUR) at 25°C and 35°C; by Van Hulle et al. (2007).

Appendix 5B: Steady state simulation results

Table S5.3. Comparison between measured data (Melse, Ploegaert et al., 2012) and steady-state simulation results for the reference case without pH control and with pH control.

Symbol		Unit	Melse et al., 2012	no pH control	pH control
η	Removal efficiency	%	82	92	94
C_{G,NH_3}^{out}	Outgoing NH3 concentration gas	ppm	2.4	1.0	0.82
$C_{BT,TN}$	Total nitrogen content buffer tank	$gN.m^{-3}$	3948	3200	3200
C_{BT,NH_3}	Total NH_3-N buffer tank	$gN.m^{-3}$	1900	1744	165
C_{BT,NO_2}	Total NO_2-N buffer tank	$gN.m^{-3}$	<10	1	0.8
C_{BT,NO_3}	Total NO_3-N buffer tank	$gN.m^{-3}$	1800	1431	3032
Q_D	Discharge rate	$m^3.d^{-1}$	0.33	0.87	0.80
RH_G^{out}	Outgoing relative humidity gas	%	>95	99.2	99.2
T_G^{out}	Outgoing temperature gas	°C	16	15	15
$T_{L,BT}$	Temperature water buffer tank	°C	16	15	15
pH	pH in water buffer tank	-	6.6	6.2	6.6

Appendix 5C: Application of a pH control system

Schematic overview of the reactions

Complete nitrification can be achieved in a biological air scrubber by applying pH control through base addition, achieving complete nitrification. An overview of the involved reactions, keeping a neutral pH, is given in Figure S5.2., comparing the scenarios without pH control and with pH control.

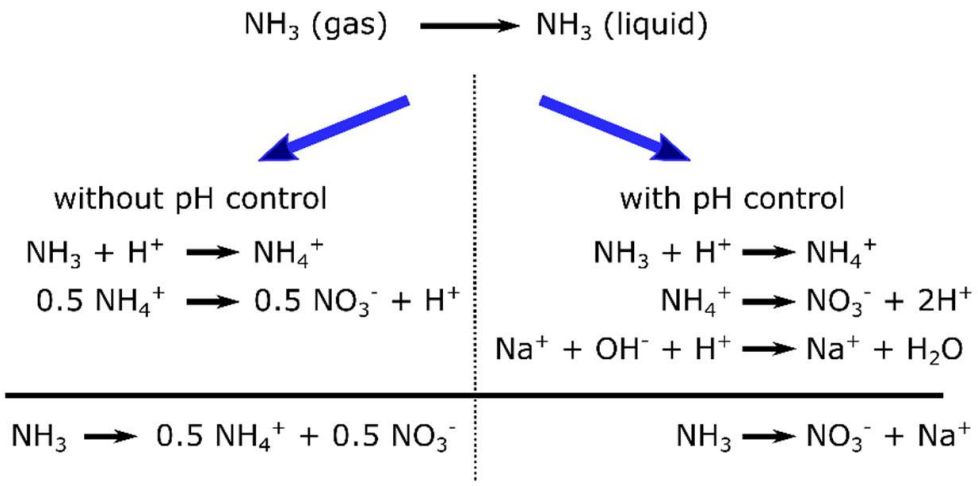

Figure S5.2. Schematic overview of the involved reaction in biological air scrubbers, taking into account that the pH does not increase or decrease. The scenarios without and with pH control are compared.

6

Long term measurement campaign at two full-scale biological air scrubbers: performance study

6.1 Abstract

This study presents the start-up and long-term performance of full-scale biological air scrubbers at a pig fattening facility in terms of ammonia removal and nitrous oxide production. Two newly-built multi-stage biological air scrubbers were continuously monitored, for these gases and for washing water characteristics. Although pH and total nitrogen in the washing water are used in practice to follow up the ammonia removal efficiency of biological air scrubbers, it was shown that the relation between these variables is not straightforward. Setting up a nitrogen balance over both air scrubbers, indicated denitrification activity. Additionally, the effect of inoculation with activated sludge of a wastewater treatment plant was investigated. The inoculated air scrubber showed a faster start-up of nitrification and less nitrite accumulation in the washing water. The ammonia removal efficiency was higher and the nitrous oxide production was slightly lower, which demonstrates the advantages of inoculation.

Van der Heyden, C., Brusselman, E., Volcke, E.I.P., Demeyer, P. (in preparation). Comparative long-term performance study of full-scale biological air scrubbers for ammonia removal.

6.2 Introduction

Biological air scrubbers or biotrickling filters are frequently used at pig housing facilities in Flanders (Belgium), the Netherlands, Germany and Denmark to lower ammonia emissions (Melse & Ogink, 2005; Van der Heyden et al., 2015). Ammonia is removed from the exhaust air by intense contact between the polluted exhaust air and the washing water, which is recirculated in the air scrubber. In the washing water, ammonia (NH_3) is in equilibrium with ammonium (NH_4^+) depending on the pH and oxidized (nitrification) to nitrite (NO_2^-) by ammonia oxidizing bacteria (AOB) and further to nitrate (NO_3^-) by nitrite oxidizing bacteria (NOB). The conversion of NH3 increases the driving force for mass transfer from the gas phase to the liquid phase.

During operation of biological air scrubbers, different operational variables are measured and controlled to allow proper functioning of the system. The pH should remain in the optimal range for microbial growth, i.e. between 6.5 and 8 (Van Hulle et al., 2007). According to the Dutch and Flemish legislation, the pH must at least remain between 6.5 and 7.5 for proper operation (InfoMil, 2015; MB31/05/2011). The total nitrogen content of the washing water is controlled by its periodic partial discharge. Electrical conductivity (EC) of the washing water is typically measured as a cheaper and easier alternative for measuring the total nitrogen content as such. In the washing water of biological air scrubbers, EC is mostly determined by the ammonium (NH_4^+), nitrite (NO_2^-) and nitrate (NO_3^-) concentrations (Melse, Ploegaert, et al., 2012). However, by only measuring EC, information about the distribution of the different nitrogen components is lost.

Although biological air scrubbers are commonly used, literature shows that the required ammonia removal efficiency of at least 70%, is not always reached (Melse, Ploegaert, et al., 2012; Van der Heyden, Brusselman, et al., 2016; Van der Heyden et al., 2015). Additionally, it is also known that biological air scrubbers have a high risk of nitrous oxide production (Dumont, Lagadec, et al., 2014; Melse & Mosquera, 2014; Melse, Ploegaert, et al., 2012; Van der Heyden, Brusselman, et al., 2016; Van der Heyden et al., 2015). However,

finding a specific reason for the low performance of biological air scrubbers both in terms of ammonia reduction and nitrous oxide production is challenging as many processes are taking place simultaneously and only few parameters are monitored (pH and EC). Most studies only provide only limited point measurements of relevant parameters, making it difficult to investigate their transient behaviour and linking it to the air scrubber performance.

Additionally, one of the disadvantages of biological air scrubbers, is the relative long start-up time, which can take several weeks or even months (Ottosen et al., 2011). Inoculating these systems by introducing nitrifying bacteria, could be a beneficial procedure to decrease this start-up time and to increase the performance. A study of Xue, Wang, et al. (2010), showed that the ammonia removal efficiency increased by 15.0%, when a biotrickling filter was inoculated with a selected consortia of AOB and NOB, originating from cattle manure solution. However, it would be more interesting to use activated sludge of nearby wastewater treatment systems, as this is freely available and easy to use, without having an additional treatment.

The aim of this study was to get more insight in the start-up and long-term performance of biological air scrubbers in terms of ammonia removal and nitrous oxide production, including the correlation between both. Therefore, two newly built multi-stage biological air scrubbers were continuously monitored, in terms of the gaseous concentrations but also the washing water characteristics, including pH, EC but also the nitrogen components, ammonium, nitrite and nitrate. It was investigated whether pH and EC gave enough information to adequately monitor and control the performance of biological air scrubbers, in terms of ammonia removal and nitrous oxide reduction. Additionally, the effect of inoculation with activated sludge of a wastewater treatment plant was investigated as cheap and easy procedure to improve the performance.

6.3 Materials and methods

6.3.1 The pig fattening facility and experimental conditions

The study was conducted from April 2016 to March 2017 at the ILVO pig fattening facility designed for experimental research, located in Flanders, Merelbeke, Belgium. This facility can house 768 fattening pigs (25 till 110 kg), divided over 16 compartments. It is mechanically ventilated via a central exhaust channel which is divided into two parallel ducts. Each exhaust duct receives air from 8 compartments, of which four have a fully slatted floor and four have a half-slatted floor, the latter reducing ammonia emissions. On average 467 animals with an average weight 67 kg were present over the measuring period. Each exhaust duct is equipped with two frequency controlled fans (Multifan, HD6, 2.4 kW, diameter: 92 cm, Vostermans Ventilation, Venlo, the Netherlands) to extract the air from the housing facility into a pressure chamber of 4 m long and through a separate biological air scrubber. The ventilation rate was controlled by temperature and pressure difference sensors inside the compartments. Both air scrubbers are designed to treat a maximum ventilation rate of 30720 $m^3.h\text{-}1$.

6.3.2 Description of the air scrubbers

Air scrubber 1 (AS1) and 2 (AS2) are respectively a three- and a two-stage crosscurrent biological air scrubber (Figure 6.1, see Table S6.1 for design specifications). Both have the same empty bed residence time (EBRT). The first stage (S1), closest to the housing facility, is generally called ‘the dust section’ since it is designed to remove dust particles. The second stage (S2) is generally called the bio section to remove ammonia. The third stage (S3), only present at air scrubber 1, is called the odour section. Both air scrubbers have a demister at the end to prevent droplets leaving the scrubber. Fresh water is added at the last stage of each air scrubber (respectively bio S2 or odour S3 section) and the washing water is discharged at the first stage (dust S1 section). The water thus flows in the opposite direction as the airflow, allowing the most concentrated washing water to come into contact with the ‘dirtiest’ air and the cleanest water with the cleanest air, increasing the driving force at the latter.

Discharge of the washing water from buffer tank AS1 S1 and AS2 S1 is automatically controlled by an EC control in the last buffer tank (respectively AS1 S3 and AS2 S2).

Both air scrubbers were started up in March 2016. Air scrubber 1 was not inoculated. Air scrubber 2 was inoculated twice over time in stage 2 with activated sludge from a nearby domestic wastewater facility. It was chosen to inoculate the two-stage air scrubber as this one was expected to have a lower performance. If the air scrubbers would perform better, this could be attributed to the inoculation. Inoculation occurred at April 4 and September 12 2016, the latter after maintenance of the air scrubbers with refreshment of the buffer tanks of the last stages (AS1 S3 and AS2 S2).

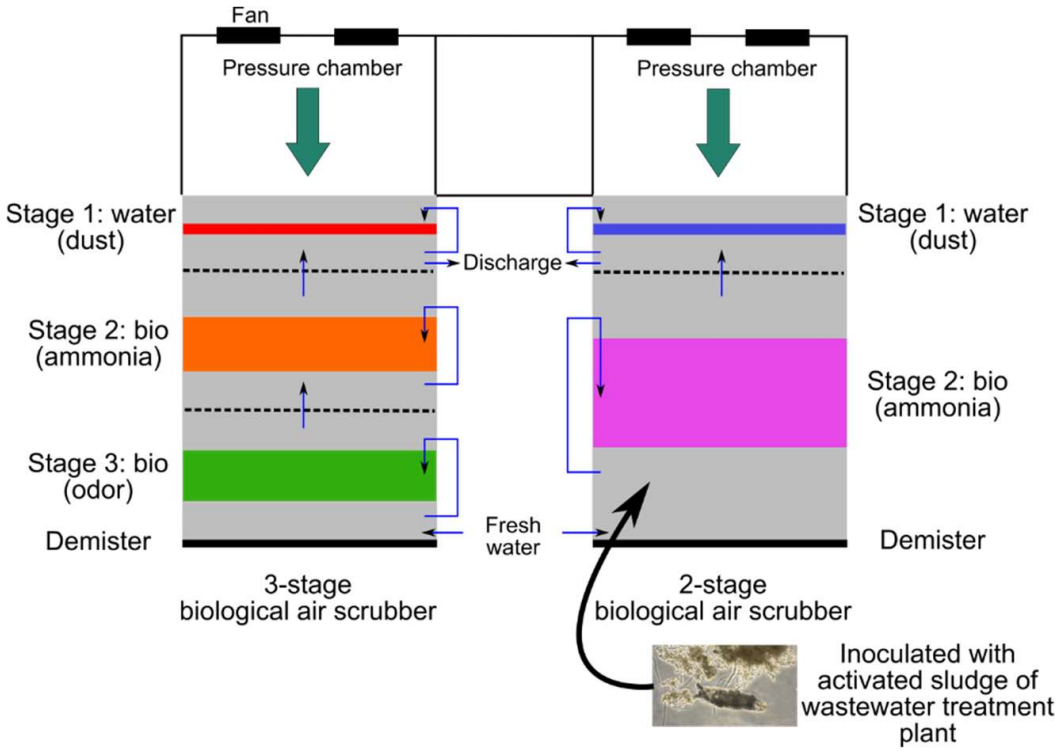

Figure 6.1. Schematic representation of the two biological air scrubbers in top view

6.3.3 Sampling strategy and measuring equipment

Gas concentration measurements of NH_3, CO_2, CH_4 and N_2O were performed using an Innova photoacoustic gas monitor 1314 connected to a CBISS multipoint sampler (LumaSense Technologies, Denmark). The analyzer was calibrated before the measurements started. Gas samples were taken

sequentially at six sampling points; one of the incoming and two of the outgoing air of each air scrubber. It was chosen to sample two points behind the scrubbers as a compromise between minimal time resolution and maximal spatial resolution. Each sampling consisted of at least 6 consecutive measurements with a 3 minutes time interval. Only the last measurement was taken into account to overcome a possible measuring delay of the gas monitor. The sampling lines in the pressure chamber were equipped with a dust filter. The sampling lines behind the scrubber were heated to 110°C to prevent condensation. Dilution by wind was avoided by using protective ducts around the sampling points (Figure 6.2). From 10 May till August, the gas analyser was not always available, resulting in periods without data.

Figure 6.2. Sampling points of the outgoing air with protective ducts and heated tubes

The gas flow rate through the air scrubber, or thus the ventilation rate of the housing facility per central exhaust duct, was calculated each minute using pressure difference measurements over the ventilation fans with a P26 differential pressure transducer (halstrup-walcher, Germany), together with the frequency of the fan, according to the fan characteristic. Ventilation rates are expressed as a percentage of the maximum ventilation rate.

Temperature and relative humidity of the incoming and outgoing air were measured every 10 minutes, using thermocouples (EE071 probe, E+E Elektronik, Germany).

The washing water of each scrubber was sampled at least every two weeks during the measuring campaign and analyzed in the lab for pH and electrical conductivity (EC). The washing water was further analysed for NH_4^+, NO_2^- and NO_3^-, using NANOCOLOR® tube tests (Table 6.1) which were analyzed with the PF-12 photometer (Machery-Nagel, Düren, Germany). When the samples were not in the measuring range of the test tube, the samples were diluted.

Table 6.1. Overview of the used NANOCOLOR® tube tests

Component	NANOCOLOR® test tube	Measuring range [mgN.L^{-1}]	Wavelength [nm]
NH_4^+	Ammonium 200	30 - 160	585
NO_2^-	Nitrite 4	0.1 - 4.0	365/385
NO_3^-	Nitrate 250	4 - 60	540

6.3.4 Calculation of process performance

The removal efficiency *RE* [%] for each pollutant was calculated on an hourly basis using the incoming concentration C_{in} [ppm] and average outgoing concentration C_{out} [ppm] of the two sampling points:

$$RE = \frac{C_{in} - C_{out}}{C_{in}} \cdot 100\% \qquad \textbf{Eq 6.1}$$

It was not necessary to recalculate the incoming and outgoing concentrations from ppm to gN.m^{-3} as the influence of the temperature difference between both on the calculation of the removal efficiency *RE* was negligible.
The loading rate *LR* [gN.h^{-1}] and removal rate *RR* [gN.h^{-1}] were calculated using the concentration and ventilation rate according to following equations:

$$LR = C_{in} \cdot \frac{M_N}{V_m} \cdot 10^{-6} \cdot Q_{vent} \qquad \textbf{Eq 6.2}$$

$$RR = \left(C_{in} - C_{out}\right) \cdot \frac{M_N}{V_m} \cdot 10^{-6} \cdot Q_{vent} \qquad \textbf{Eq 6.3}$$

with M_N the molecular weight of nitrogen [gN.mol^{-1}], V_m the molecular volume [m^3.mol^{-3}], based on the prevailing temperature and pressure and Q_{vent} the ventilation rate [m^3.h^{-1}].

A nitrogen mass balance was calculated on an hourly basis. The incoming and outgoing mass of ammonia and nitrous oxide was calculated according Eq 6.2. The total nitrogen in the washing water was calculated from the hourly EC measurements. Accumulation of nitrogen in the buffer tank was calculated as the difference between two time instances, which was zero as the EC was constant. The discharge of nitrogen from the buffer tank was calculated by multiplying the discharge volume with the total nitrogen concentration at the moment of discharge.

6.3.5 Statistical analysis

The statistical analyses were carried out using the SAS® statistical software (SAS Institute Inc, Cary, USA). A one-way ANOVA was used to compare the ammonia removal and nitrous oxide production between the two biological air scrubbers for different periods. Linear regression analysis was used to compare the relationship between the performance of the air scrubbers in terms of ammonia removal and nitrous oxide production with the washing water parameters FA and FNA. All comparisons and regressions were assessed at the 95% (p 0.05) confidence level.

6.4 Results and discussion

6.4.1 General characteristics and air scrubber performance

6.4.1.1 *Air inlet characteristics*

An overview of the air inlet characteristics, i.e. ventilation rate, temperature, relative humidity and incoming concentrations and load of NH_3 and N_2O is presented in Figure S6.1 (Appendix 6B).

The ventilation rate was relatively constant for both air scrubbers, with an average of 50.5 ± 5.0% at AS1 and 52.2 ± 7.3% at AS2 (Figure S6.1 A). Some

peaks occurred when the ventilation was manually switched to 100%. The maximum ventilation rate measured 76.8% (AS1) and 82.6% (AS2). The ventilation system was mostly working at half of its capacity and is thus slightly over-dimensioned. The daily profile shows, on average, a 10% higher ventilation during the day compared to the night. On average, the incoming temperature was 20.5 ± 4.4°C at AS1 and 19.9 ± 4.0°C at AS2 (Figure S6.1 B). Due to the change in latent and sensible heat, the outgoing temperature was approximately 5°C lower (15.6 ± 4.5°C at AS1 and 14.9 ± 4.2°C at AS2), which is consistent with other studies (Melse, Ploegaert et al., 2012; Van der Heyden et al., 2016). The relative humidity of the incoming air was on average 67 ± 9% at AS1 and 67 ± 7% at AS2 and was always >95% at the outgoing air of both air scrubbers.

The average incoming NH_3 concentration measured 10.0 ± 2.6 ppm for AS1 and 11.4 ± 4.8 ppm for AS2 over the entire measurement period. The maximum incoming NH_3 concentration in AS1 was 20 ppm. AS2 had a higher incoming concentration from January till March 2017, with maximum values of 35 ppm. This was due to some newly introduced pigs, which showed a different dunging behaviour as they dunged at the solid floored lying area instead of the slatted dunging area, resulting in more dirty compartments and higher emissions of ammonia from the surfaces. The average incoming ammonia load was 91 ± 24 $gN.h^{-1}$ for AS1 and was slightly higher for AS2, i.e. 106 ± 46 $gN.h^{-1}$. The incoming N_2O concentration was on average 0.39 ± 0.13 ppm for AS1 and 0.40 ± 0.13 ppm for AS2, with an average load of 9 $gN.h^{-1}$ for both air scrubbers.

It can be concluded that the inlet air conditions were comparable for both air scrubbers. This is important since ventilation rate, temperature and incoming concentrations can have a significant effect on the ammonia removal efficiency (Melse, Ploegaert et al., 2012).

6.4.1.2 *Ammonia removal and nitrous oxide production*

The measurement results in terms of ammonia removal efficiency and nitrous oxide production at both air scrubbers are summarized in Figure 6.3 (A, B and

C). Although the incoming ammonia concentrations were comparable for both air scrubbers, with slightly higher incoming concentrations at AS2 (Figure S6.1C, Appendix 6B), the outgoing ammonia concentration was on average lower for AS2 (2.8 ± 2.4 ppm) than for AS1 (4.6 ± 2.6 ppm). The average ammonia removal efficiency was 50.5 ± 29.7% for AS1, which was significantly lower ($p < 0.0001$) than the removal efficiency of AS2 (70.0 ± 27.4%). Figure 6.3A shows that the ammonia removal efficiency varied significantly during the entire measuring period, both for the two-stage and three-stage air scrubber. After start-up during the first month, the average ammonia removal efficiency measured only 11.1 ± 6.9% for AS1 and 23.7 ± 9.4% for AS2. The available data shows that the ammonia removal efficiency only reaches the required 70% after 2 months. However, this level could not always be maintained over the entire measurement period. Despite some missing data during the start-up period, the results show the slow start-up of both air scrubbers in terms of ammonia removal.

The lowest ammonia removal efficiency measured was -408% for AS1 and -17% for AS2, meaning that ammonia was produced in the air scrubbers. This occurred at September 8th and 9th when the air scrubbers were switched off for maintenance, while the ventilation was still running. The maximum ammonia removal efficiency measured 92.45% for AS1, which occurred at January 9th at midnight and was 99.12% for AS2, which occurred at November 21st, also during the night. Taking into account only the last period, from January 2017 onwards, when the performance was more stable, AS1 reached an average ammonia removal efficiency of 77.3 ± 8.0% and AS2 89.6 ± 6.2% ($p < 0.0001$). Thus, even with a higher incoming ammonia load, the two-stage air scrubber performed better in terms of ammonia removal.

Considering nitrous oxide, the outgoing concentration was 0.53 ± 0.15 ppm at AS1 and 0.52 ± 0.16 ppm at AS2 (Figure S6.1D, Appendix 6B). Nitrous oxide is thus produced instead of removed, and is therefore expressed as a production (negative removal efficiency). Although the outgoing concentrations are low, nitrous oxide production should be avoided at all times. The production varied significantly during the entire measuring period for both scrubbers (Figure

6.3B). The average nitrous oxide production over the entire measuring period was 39.1 ± 28.5% for the three-stage air scrubber and 31.0 ± 23.9% for the two-stage air scrubber. Taking into account only the last period, from January 2017 onwards, when the ammonia removal efficiency reached a more constant level, the nitrous oxide production measured 48.3 ± 28.3% at AS1 and 29.2 ± 17.9% at AS2. This is slightly higher at AS1 ($p < 0.0001$). The highest production measured was 92.5% for AS1 and 99.1% for AS2, meaning almost a doubling compared to the incoming concentration.

When the relative nitrous oxide production is expressed as the produced nitrous oxide concentration compared to the incoming ammonia in the air scrubber, the average of the three-stage air scrubber amounts 5.3 ± 1.4% and of the two-stage air scrubber 4.7 ± 1.5%. This is within the boundaries found in other biological air scrubbers, ranging around 3% up to 66% (Melse, Ploegaert, et al., 2012; Van der Heyden, Brusselman, et al., 2016). The highest production was denoted when an extra denitrification tanks were present. These values are high compared to municipal wastewater treatment plants applying the nitrification-denitrification system, where this rate varies between 0 and 15% (Daelman et al., 2013; Kampschreur et al., 2009; Yoshida et al., 2014). Although nitrification was very low at start-up, the N_2O production expressed to incoming nitrogen amounted already 4.1 ± 0.8% and 3.9 ± 0.8% for AS1 and AS2, respectively. One very large peak can be observed, up to 20%, for both air scrubbers at November 10th. This is caused by a large incoming nitrous oxide concentration, up to almost 3.6 and 2.3 ppm at AS1 and AS2 respectively. At that moment, the outgoing nitrous oxide concentrations were lower than the incoming concentration, i.e. 2.8 at AS1 and 2.2 ppm at AS2. No clear explanation could be given for this peak.

6.4.1.3 *Washing water characteristics*

Figure 6.3 (D, E, F and G) summarizes the measurement results for the washing water of both air scrubbers, in terms of pH, EC, the nitrogen components TAN, NO_2^- and NO_3^- expressed to the total nitrogen concentration as sum of the three components, as well as FA and FNA. In Figure S6.2 (Appendix 6B), the concentrations of the nitrogen components are given as such.

By considering the washing water analysis, more insights in the process operation could be gathered. The pH of the washing water in both air scrubbers was high at start-up with values above 8.0 (Figure 6.3D). This high pH is due to ammonia absorption, while nitrification is not yet occurring or high enough to reduce the pH. At that moment, only TAN is present in the washing water (Figure 6.3E). A study of Xue et al. (2010) showed that the pH increased from 7.04 to 8.81 when ammonia was transferred from the gas phase to the liquid phase and oxidation of ammonia did not yet occur. When nitrification started, the relative amount of TAN decreased and of NO_2^- and NO_3^- increased (Figure 6.3E), while the pH decreased quite rapidly to a constant value around 7 to 7.5 (Figure 6.3D). This pH drop can be explained by a net production of protons since absorption of an NH_3 molecule and conversion into NH_4^+ uses one proton, whereas nitrification of one NH_4^+ molecule produces two protons. As a result, once half of the ammonium has been converted into nitrite by the bacteria, the pH decreases, which inhibits further ammonia conversion. Furthermore, a constant value of about 50% TAN was indeed observed in the two air scrubbers buffer tanks, despite the different total nitrogen content in the water (Figure 2E). This incomplete nitrification has also been observed in other studies (Melse & Mosquera, 2014; Mosquera et al., 2011; Ottosen et al., 2011; Van der Heyden, Brusselman, et al., 2016).

During the entire measurement campaign, different EC set points were applied, resulting in varying EC values and discharge rates. The EC values went up in the first buffer tank to 60 $mS.cm^{-1}$ at the three-stage air scrubber and to 65 $mS.cm^{-1}$ at the two-stage air scrubber (Figure 6.3E). These values are rather high compared to the normal practices but were chosen to test both air scrubbers in more extreme conditions.

It can be observed that the three-stage air scrubber showed more fluctuation in EC compared to the two-stage air scrubber. Keeping the three buffer tanks of AS1 at the desired EC level was encountered difficult to control. The EC setpoint is measured in the last buffer tank (S3), while most ammonia is absorbed in the first sections (S1 and S2). Hereby, the EC of these two latter stages increases faster than for S3, and this does not result in an increased

discharge rate. The discharge rate fluctuated a lot between periods with high discharge rate and longer periods where discharge was reduced to zero. In the two-stage air scrubber, the discharge rate was more constant. The discharge rate measured on average 0.44 $m^3.day^{-1}$ for AS1 and 0.30 $m^3.day^{-1}$ for AS2. Therefore the hydraulic retention time (HRT) of the water in the buffer tank was respectively 7.3 and 7.7 days, meaning it took approximately 7 days before all water was refreshed in the buffer tank or 13% of the buffer tank was refreshed per day. Most biological air scrubbers installed at pig housing facilities have a comparable HRT and it can even be longer as the buffer tank can be very large.

Due to the high EC values, the concentrations of NH_4^+, NO_2^- and NO_3^- were high as well, reaching values up to 7.9, 6.4 and 1.5 $gN.L^{-1}$, respectively at AS1 and 9.7, 3.0 and 4.6 $gN.L^{-1}$, respectively at AS2 (Figure S6.2, Appendix 6B). From Figure 6.3E it can be observed that the three-stage air scrubber had a significantly higher nitrite concentration and lower nitrate concentration compared to the two-stage air scrubber. Additionally, shifts between NO_2^- and NO_3^- occurred at AS2. The free ammonia (FA) and free nitrous acid (FNA) concentration of the washing water, which is determined by the concentration of TAN and TNO2 and the pH, are the most relevant parameters as they are the substrate but can also have inhibitory effects (Ottosen et al., 2011). Inhibition of AOB (*Nitrosomonas*) and NOB (*Nitrobacter*) caused by FA occurs from 10 to 150 $mgN.L^{-1}$ and 0.1 to 1.0 $mgN.L^{-1}$, respectively, whereas FNA concentrations inhibit both bacteria at 0.22 to 2.8 $mgN.L^{-1}$ (Anthonisen et al., 1976). The FA concentration varied between 0 and 119 $mgN.L^{-1}$ in S1 of AS1 and between 0 and 90 $mgN.L^{-1}$ in S1 of AS2 (Figure 6.3F). The FNA concentration increased up to 1.96 and 0.66 $mgN.L^{-1}$ in S1 for the three-stage and two-stage air scrubber, respectively (Figure 6.3G). In the two-stage air scrubber, the FNA concentration remained zero at the end of the measuring period, as nitrite was fully converted to nitrate. The FA and FNA concentration in both air scrubbers, far exceeded the boundary for inhibition of NOB, which could explain the observed nitrite accumulation. Accumulation of nitrite in

biological air scrubbers is a frequent problem (Melse, Ploegaert, et al., 2012; Ottosen et al., 2011; Van der Heyden, Brusselman, et al., 2016).

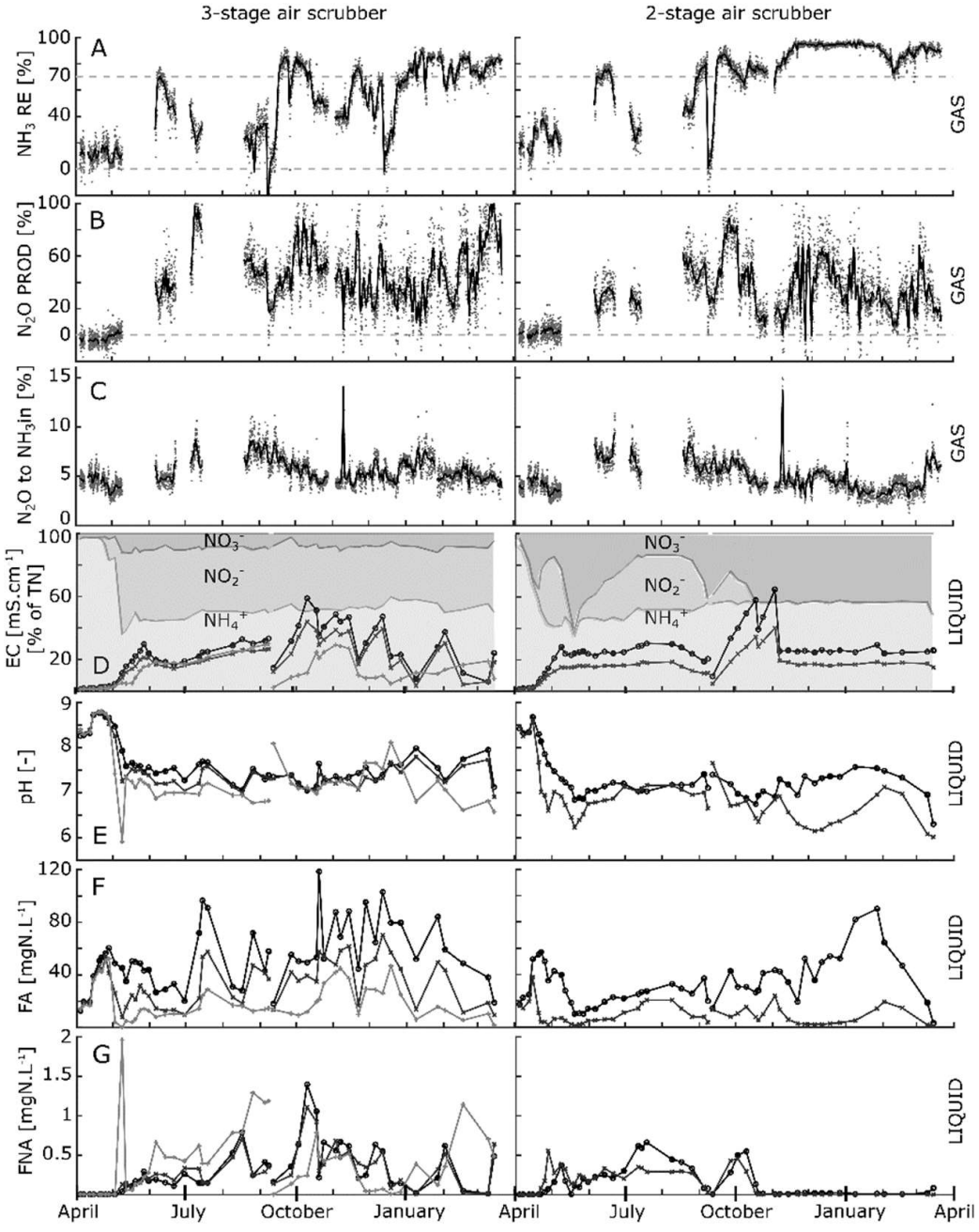

Figure 6.3. Overview of the performance in terms of ammonia removal efficiency, nitrous oxide production and the washing water conditions: EC, pH and the composition of the nitrogen components. The two-stage air scrubber was inoculated at April 4th and September 12th 2016.

6.4.2 Estimation of the total dissolved nitrogen content of the washing water using EC data

Figure 6.4 shows the electrical conductivity (EC) in function of the total dissolved nitrogen components, expressed as sum of NH_4^+, NO_2^- and NO_3^-, in the washing water of both air scrubbers. A correlation between both exists as the electrical conductivity is a measure for the total ions in the water. It is assumed that ammonium, nitrite and nitrate are the most prevalent ions in air scrubbers (Melse, Ploegaert, et al., 2012). Based on the molar ionic conductivity of each component (Lide, 2005), the electrical conductivity of the ammonium, nitrite and nitrate ions equal 5.25, 5.13 and 5.10 $(mS.cm^{-1}).(gN.L^{-1})^{-1}$ in an ideal solution. Taking into account that 50% of the ions are ammonium and the other half is nitrite or nitrate (both assumed to be 25%), the weighed average is 5.18 $(mS.cm^{-1}).(gN.L^{-1})^{-1}$. The same value was used by Ottosen et al. (2011) to make an estimation of the total nitrogen content in his experiments based on a measured EC. However, this is only valid in ideal solutions. A correlation based on experimental data would be more reliable for practical conditions.

Therefore, a linear regression based on measurements in this study was introduced. There was no significant difference between the different stages of the air scrubbers and between both air scrubbers, allowing to set up one linear regression using all 250 data points (Figure 6.4). The obtained linear regressing showed a lower slope than the one based on the molar ion conductivity. For total dissolved nitrogen concentrations below 4 $gN.L^{-1}$, the linear regression based on the molar ion conductivity fits well for the experimental results in this study, but more deviation is found for higher concentration. This can be attributed to non-idealities taking place in concentrated solutions, which should be characterized by activities instead of concentrations.

Two other correlations between EC and the nitrogen components in the washing water of biological air scrubbers treating pig exhaust air were found in literature. Melse, Ploegaert et al., (2012) set up a correlation based on one-year measurements at one biological air scrubber, for total nitrogen

concentrations between 2 and 6 gN.L^{-1}. Lagadec et al. (2015) studied 31 different biological air scrubbers and also found a very strong correlation (R^2 = 0.981) between EC and ammoniacal nitrogen in the water. To be able to compare this correlation with the one of Melse, Ploegaert et al (2012) and the one in this study, it was assumed that 50% of the total nitrogen in the washing water was present as ammonium, resulting in a correlation between EC and total nitrogen. Comparison of the different correlations shows that these based on experiments have a lower slope compared to the one based on molar ion conductivity in an ideal solution. This is especially pronounced at higher total nitrogen concentrations. The biggest difference was found for this study, i.e. a difference of approximately 30% at a total nitrogen concentration of 18 gN.L^{-1}. This could be due to the higher concentrations, diverging more from the ideal solution or because of the presence of other ions in the washing water (not tested).

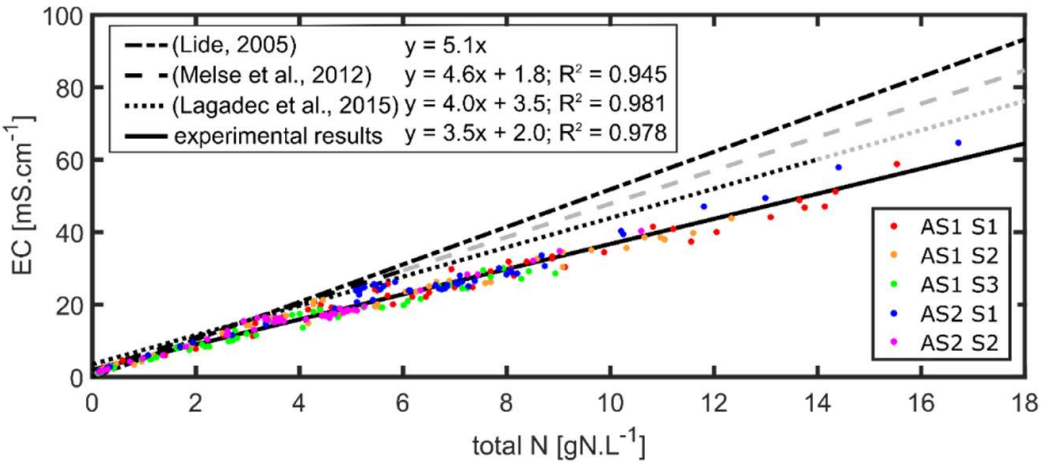

Figure 6.4. Electrical conductivity (EC) in function of the total dissolved nitrogen compounds (total N) in the recirculation water. Dots represent measurements of each buffer tank of both air scrubbers. Straight lines represent the corresponding linear regressions (black: inside the measurement range; grey: outside the measurement range).

These correlations are very useful to estimate the total nitrogen content in the washing water of air scrubbers, when only EC measurements are available. At lower electrical conductivity, all correlations show comparable results. However, at higher EC, greater differences occur between the correlation based on molar electrical conductivity and the measurements. The correlation in this study was validated in more extreme conditions, moreover, it includes

the other correlations, and can therefore be used to estimate the total nitrogen content at biological air scrubbers.

6.4.3 Estimation of denitrification

A nitrogen mass balance was calculated considering the nitrogen fluxes of the inlet air, outlet air, discharge water and accumulation of dissolved nitrogen compounds in the washing water of the buffer tank to determine the fate of the incoming nitrogen and to check if all nitrogen was recovered with the gas and liquid measurements.

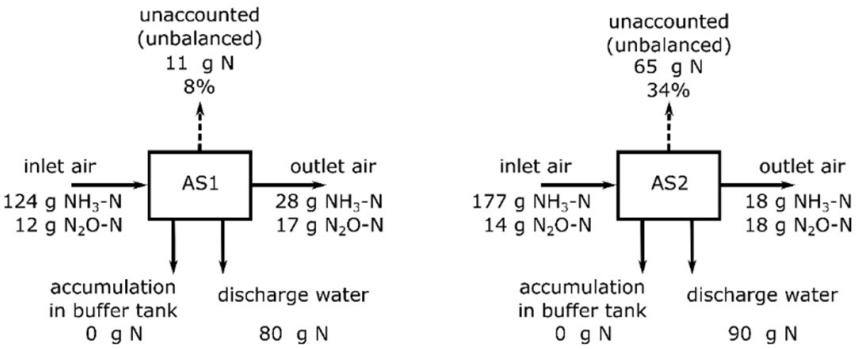

Figure 6.5. Daily averaged unaccounted nitrogen to the total incoming nitrogen expressed in percentage (A); Overall fluxes of the nitrogen compounds on hourly basis averaged over the period January till March 2016 (B)

Figure 6.5 shows the overall fluxes of the nitrogen compounds on an hourly basis [$gN.h^{-1}$], averaged over the period January till March 2016, when the operation can be more considered to be in steady-state. The ammonia flux of the inlet air was on average of 124 and 177 g NH_3-$N.h^{-1}$ for the three-stage and two-stage air scrubber, respectively. The outgoing ammonia flux was on average 28 and 18 g NH_3-$N.h^{-1}$. The removal rate thus amounted to 96 and 159 g NH_3-$N.h^{-1}$ and the ammonia removal efficiency was 77% and 89% for AS1 and AS2, respectively. A significantly higher ammonia removal efficiency in the two-stage air scrubber was thus noted. As found before, the production of nitrous oxide was more comparable, having an average production rate of 5 and 4 $gN.h^{-1}$ for AS1 and AS2, respectively. The removed nitrogen was expected to up in the washing water, either as accumulated or as discharged nitrogen. It can be noted that for both air scrubbers, accumulation in the washing water equals on average zero. This means that all nitrogen that was

absorbed, was discharged. Still, on average 11 g N and 65 g N, corresponding 8% and 34% of the incoming nitrogen could not be balanced for the three-stage and two-stage air scrubber, respectively.

This nitrogen imbalance, which was also found in other studies (Blázquez et al., 2017; Melse, Ploegaert, et al., 2012), could be due to measurement errors or nitrogen sinks such as denitrification processes or emissions not yet accounted for. Estelles, Calvet, Melse, & Ogink (2012) indicated the airflow measurement as the largest source of errors in the nitrogen balance. According to Melse, Ploegaert, et al. (2012), some nitrogen might have accumulated on the packing material or incorporated in the biomass, or might have been emitted as nitrous oxide (N_2O), nitrogen gas (N_2) or NO_x. Since nitrous oxide and the washing water characteristics were measured more continuously in this study, denitrification was hypothesized to explain the gap in the nitrogen balance (N_2). The presence of denitrifiers in the biofilm of the biological air scrubber was confirmed through molecular analysis (Chapter 7) and by other studies (Blázquez et al., 2017).

6.4.4 Relation between ammonia removal efficiency and washing water characteristics

Ammonia removal is not measured continuously at air scrubbers but the performance is followed up by monitoring the washing water characteristics like EC and pH. It is assumed that keeping these variables in between the desired ranges gives enough guarantee for proper operation of the air scrubbers.

The pH of the washing water must remain between 6.5 and 7.5, according regulation in Flanders, the Netherlands and Germany (DLG, 2014; InfoMil, 2013; MB31/05/2011). Nitrification has a pH optimum at 7.23 (Van Hulle et al., 2007). A pH above 7.5 is disadvantageous for the ammonia removal efficiency since this shifts the equilibrium in the washing water towards ammonia, reducing the mass transfer driving force from gas to liquid. Although a lower pH is more beneficial for ammonia mass transfer, it is undesirable for nitrification as it will result in inhibitive FNA concentrations in the washing

water. A low pH of the washing water concerns as well a risk for corrosion and for nitrous gases formation (DLG, 2014). Additionally, the risk to strip HNO2 (g) from the washing water exist, especially when high nitrite concentrations are present. From preliminary research results in the Netherlands, it appears that high concentrations of nitrogen oxides are measured in the exhaust air of biological air scrubber having a low washing water pH (Melse, 2017; and personal communication).

EC is the most important control parameter in biological air scrubbers as it is a measure for the total nitrogen content, the discharge rate and the HRT. By setting the nitrogen content, also the nitrogen components are set as 50% of the total nitrogen in the washing water is ammoniacal nitrogen and the other 50% can be either total nitrite or nitrate. In Flanders, the total nitrogen content may not exceed 3.2 $gN.L^{-1}$, thereby keeping the FA and FNA concentrations below the inhibitory levels at pH between 6.5 and 7.5 (maximum FA = 13.7 $mgN.L^{-1}$; maximum FNA = 1.5 $mgN.L^{-1}$).

FA has a direct impact on the mass transfer and FA and FNA are substrates for, but can also inhibit, the nitrifying bacteria. Figure 6.6 and Figure 6.7 show the ammonia removal efficiency and nitrous oxide production in terms of the FA and FNA concentrations, without considering the start-up period. The results of the linear regression analysis can be found in Appendix 6C (Table S6.2). The ammonia removal efficiency showed a negative correlation with the FA concentration, especially for the last buffer tank (Figure 6.6; AS1 S3: R^2 = 0.82 and AS2 S2: R^2 = 0.36). This could be assigned to the mass transfer, which decreased at higher ammonia concentrations in the washing water. The FNA concentration did not show a significant correlation with the ammonia removal efficiency in AS1. In AS2 however, a weak correlation (R^2 = 0.15) was found (Figure 6.7), for which a higher FNA concentration resulted in a lower ammonia removal efficiency.

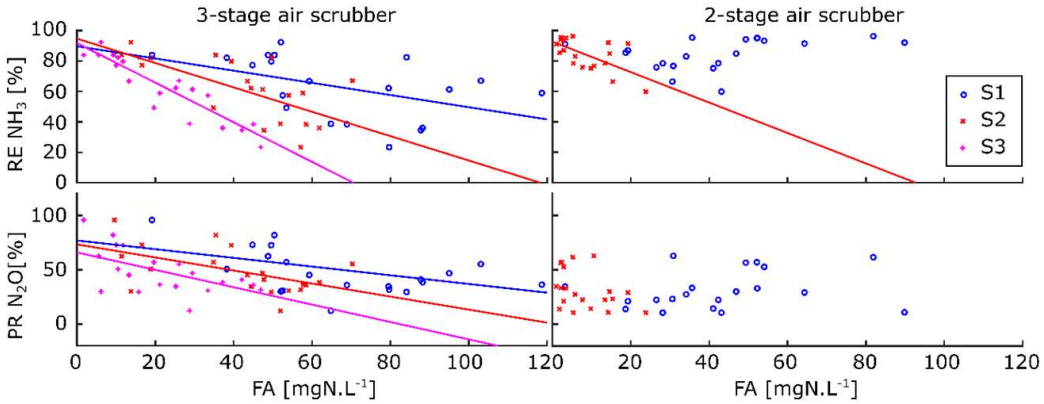

Figure 6.6. Ammonia removal efficiency and nitrous oxide production compared to the free ammonia concentration (FA) in the washing water at each stage.

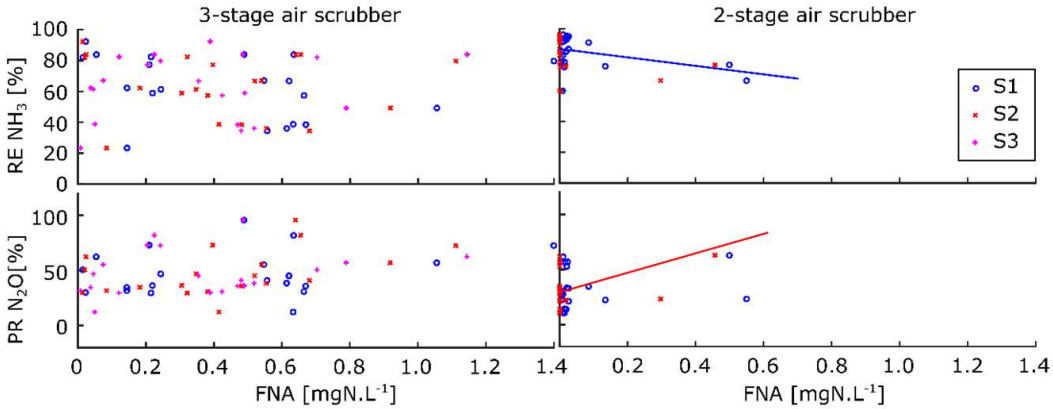

Figure 6.7. Ammonia removal efficiency and nitrous oxide production compared to the free nitrous acid concentration (FNA) in the washing water at each stage.

It was further investigated if the assumption of keeping pH and total nitrogen (TN) in between the desired ranges, gives enough guarantee for reaching an ammonia removal efficiency of at least 70%. Figure 6.8 summarizes the measurements of pH and TN in the washing water of the last buffer tank of both air scrubbers (S3 and S2, respectively), in terms of the ammonia removal efficiency in the gas phase from October till March. The start-up period was not taken into account. The Flemish boundaries for pH (6.5 and 7.5) and for TN (3.2 $gN.L^{-1}$) were considered. For AS1, 86% of the measured efficiencies corresponded well with the respective (Figure 6.8) pH and TN settings. So when the washing water pH remained in between 6.5 and 7.5 and TN remained below 3.2 $gN.L^{-1}$, an ammonia removal efficiency above 70% was reached, while outside these boundaries, the ammonia removal efficiency was less than 70%. Only one of the 21 measurements was false positive (5%), as the 70% ammonia removal efficiency was not reached, while the pH and TN values

were between the boundaries. However, during two measurements (10%) the ammonia removal efficiency was above 70% while the boundaries were exceeded. For AS2, the latter was even more pronounced as 90% of the measurements were outside the boundaries and still more than 70% ammonia removal efficiency was reached. This means that keeping the washing water pH and TN in between the imposed boundaries gives a higher change of reaching a good air scrubber performance in terms of ammonia removal but it is not necessarily bad when the boundaries are exceeded, especially for TN. It could be considered to elevate the maximum limit of 3.2 $gN.L^{-1}$ in Flanders or even to omit this boundary. Allowing higher TN values would reduce operation costs as less discharge would be necessary. However, these results also show that it is difficult to follow up the ammonia removal efficiency just be measuring pH and TN in the washing water. A direct measurement of the ammonia removal efficiency by measuring incoming and outgoing ammonia concentrations in the gas phase would therefore be recommendable.

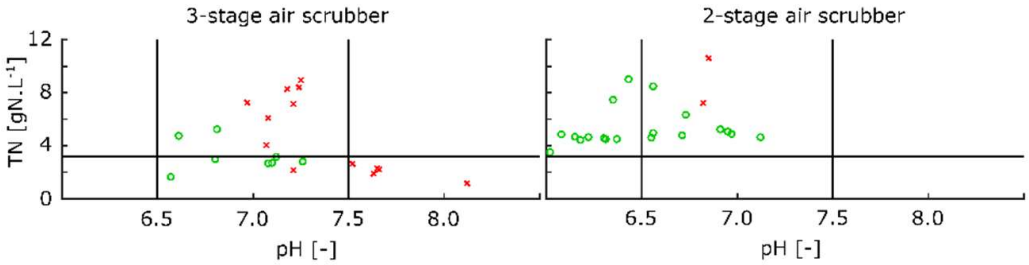

Figure 6.8. Measurements of pH and TN in the washing water of the last buffer tank of both air scrubbers (S3 and S2, respectively), linked with the ammonia removal efficiency in the gas phase from October till March. The start-up period was not taken into account. The Flemish boundaries for pH (6.5 and 7.5) and for TN (3.2 $gN.L^{-1}$) are displayed. An ammonia removal efficiency above 70% is indicated in green, below 70% in red.

6.4.5 Nitrous oxide production in biotrickling filters

It could be expected that ammonia removal and nitrous oxide production are positively correlated as nitrous oxide is a side product of the nitrification reactions. Figure 6.9 shows the correlation between the ammonia removal rate and the nitrous oxide production rate, with a correlation coefficient (r) at AS1 and AS2 of 0.11 ($p < 0.001$) and 0.30 ($p < 0.001$), respectively. A positive correlation between both thus exists, but is rather low. Some inexplicable outliers in the nitrous oxide production rate can be observed, decreasing the

correlation coefficient significantly. Correlating the nitrous oxide production with the ammonia oxidation rate, thus the ammonia that is converted during nitrification, is considered more correct. However, the ammonia oxidation rate could not be calculated from the available data.

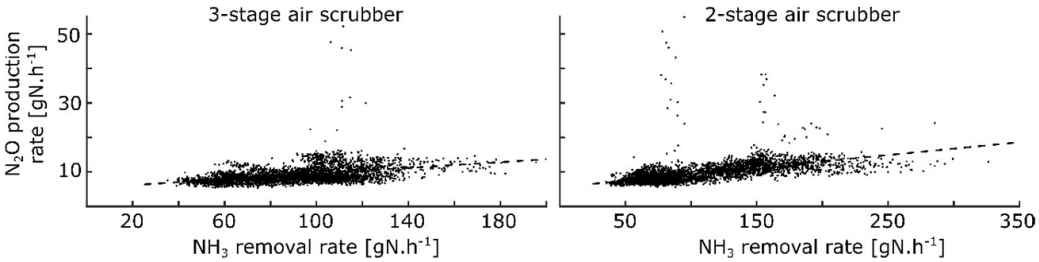

Figure 6.9. Correlation between ammonia removal rate [gN.h^{-1}] and nitrous oxide production rate [gN.h^{-1}] at the two air scrubbers.

For the three-stage air scrubber, a negative linear regression could be found between FA and the nitrous oxide production (Figure 6.6), but the R^2 value was less than 0.30 and this trend was not found for the two-stage air scrubber. Correlating the FNA concentration with the nitrous oxide production was only significant for AS2 S2 (Figure 6.7). Only a few FNA concentration values were higher than zero for this air scrubber, thus results should be considered carefully.

Nitrous oxide production in wastewater treatment processes have been studied extensively over the last decades. Based on the current knowledge, three main microbial pathways leading to N_2O emissions are put forward, namely hydroxylamine (NH_2OH) oxidation and nitrifier denitrification, both carried out by ammonia oxidizing bacteria (AOB), besides heterotrophic denitrification by heterotrophic bacteria (Kampschreur et al., 2009; Wunderlin et al., 2013, 2012). The two AOB pathways are considered to be the major contributors in nitrification processes and can occur simultaneously (Massara et al., 2017). Nitrous oxide production via hydroxylamine oxidation is favoured at high ammonia and low nitrite concentrations, in combination with high metabolic activity of AOB (at 2 to 3 mg $O_2.L^{-1}$). The contribution of nitrifier denitrification by AOB increases at higher nitrite and lower ammonia concentrations, or at oxygen-limiting conditions (Wunderlin et al., 2012). Heterotrophic denitrification normally serves as a mechanism of nitrous oxide

consumption but can be disturbed and stopped at the N_2O formation stage (Massara et al., 2017) due to oxygen inhibition, nitrite accumulation or a limited availability of biodegradable organic compounds (Wunderlin et al., 2012). Several possible conditions, which mostly prevail in biological air scrubbers, can thus be identified to favour nitrous oxide production: low dissolved oxygen concentration, accumulation of nitrite, rapidly changing (dynamic) conditions such as pH and concentration fluctuations in the washing water and a low ratio of COD to N-compounds during heterotrophic denitrification.

The effect of nitrite accumulation on ammonia removal and nitrous oxide production as such, was also be assessed. As the total nitrogen concentration can also have a negative effect, two periods with the same EC but with and without nitrite accumulation were compared in the two-stage biological air scrubber. From October 1st till October 15th, the air scrubber shows a significant nitrite accumulation (11% of the total nitrogen content or 0.6 gN.L-1, at an average EC of 29 mS.cm^{-1}) (Figure 6.3). From October 15st till October 31th, nitrite was no longer present (0% of the total nitrogen content or <0.01 gN.L^{-1}, at an average EC of 34 mS.cm^{-1}). During October, the ammonia removal efficiency was comparable, reaching on average 75% during both periods. However, the nitrous oxide production was lower when there was no nitrite accumulation in the washing water (22 ± 18%) compared to the period with nitrite accumulation (51 ± 22%). Nitrite accumulation could thus have a negative effect on the nitrous oxide production, as also found in wastewater treatment plants. However, during the following months, the nitrous oxide production increased again, even when nitrite was not present, and can thus not be considered to be the only reason for nitrous oxide production. Both nitrifier denitrification or heterotrophic denitrification occur at high nitrite concentrations. From the mass balance, denitrification is expected, making the latter pathway considerable. Additionally, nitrifier denitrification normally occurs at low ammonia concentrations, which are conditions which are mostly not found in biological air scrubbers. Therefore, it is assumed in this study, that the main nitrous oxide production pathway is heterotrophic denitrification.

6.4.6 Effect of inoculation

At start-up, 100% of the total nitrogen was made up of ammonium, which was converted to nitrite and/or nitrate when nitrification starts, shown in Figure 6.3. For the three-stage non-inoculated air scrubber, the decrease in the percentage of TAN to the total nitrogen started after 15 days. For the two-stage inoculated air scrubber, an immediate decrease in TAN was observed, meaning that nitrification occurred immediately. This is due to the immediate presence of AOB and NOB from the activated sludge, which can right away start nitrifying. After 18 days for the inoculated two-stage air scrubber and 30 days in the non-inoculated three-stage air scrubber, a steady state is reached whereby 50% ammonium remained in the washing water, despite the changing EC. However, it cannot be stated from this study, if this faster nitrification also results in a faster reaching of the required 70% ammonia removal as this data was not available.

In the non-inoculated air scrubber, a large part of ammonium is immediately converted to nitrite and not further to nitrate, as only 10% of the total nitrogen during the whole measurement campaign was nitrate and 40% remained in the washing water as nitrite (Figure 6.3). In the inoculated air scrubber, some shifts between nitrite and nitrate are observed but afterwards full nitrification towards nitrate was reached. It can be reasonably assumed, that in the inoculated air scrubbers, NOB are present from the beginning, making it easier for them to compete with AOB, thereby reducing nitrite accumulation.

Concerning the performance of both air scrubber, the inoculated air scrubber showed a higher ammonia removal efficiency, both taking into account the total measurement period and only the steady-state period, showing for the latter an increase of 12% (77% at AS1 and 90% at AS2). Additionally, a lower average nitrous oxide production was observed at the inoculated air scrubber (48% at AS1 and 29% at AS2). However, it is difficult to compare both air scrubbers just by terms of inoculation as also some other aspects of the system differ, such as number of stages, liquid to gas ratio, as well as the loading rate because not exactly the same air enters the two air scrubbers. The study of Xue et al. (2010) showed that inoculation could enhance the ammonia removal

efficiency with 15% compared to a situation without inoculation but inoculation occurred inside the same biotrickling filter and thus also the effect of time could possibly explain this higher removal efficiency. Despite the promising results concerning inoculation of biological air scrubber, it thus remains difficult to concluded that inoculation of biological air scrubbers with activated sludge from a wastewater treatment plant, will enhance their performance. Although, nitrification started faster in the inoculated air scrubber and less nitrite accumulation occurred. As inoculation is an easy and cheap method, it can still be recommended to inoculate biological air scrubbers in practice to benefit from these advantages.

6.5 Conclusions

- The performance of two full-scale biotrickling filters for ammonia removal was monitored over a long term (355 days) based on gas phase and liquid phase measurements.
- Using the extensive analysis of the washing water of both air scrubbers, a correlation between electrical conductivity (EC) and total nitrogen (TN) concentration as sum of the ions ammonium, nitrite and nitrate was set up and compared to other correlations in literature. The correlation was confirmed at higher total nitrogen concentration and is therefore accurate to calculate TN based on EC measurements at any biological air scrubber.
- The nitrogen balance at the three-stage and two-stage biological air scrubber showed a large imbalance of 8% and 34%. Since nitrous oxide and the washing water characteristics were measured continuously in this study, it was hypothesized that the unaccounted nitrogen is emitted as N_2 by denitrification.. This was confirmed by the presence of denitrifying bacteria.
- The ammonia removal efficiency after start-up was on average 77% and 90% for the three-stage and two-stage air scrubber, respectively. In practice, the air scrubber performance is not monitored by ammonia measurements but through the washing water characteristics EC and pH. It was shown that keeping these variables in between the desired ranges

gives enough guarantee for proper operation of the air scrubbers but exceeding these boundaries does not necessarily imply that the ammonia removal efficiency is not reached.

- A high production of nitrous oxide compared to the incoming concentration was observed, on average 48% and 29%, after start-up of the three-stage and two-stage air scrubber, respectively. Around 5% of the incoming ammonia was emitted as the strong greenhouse gas, nitrous oxide. The ammonia removal rate was correlated with the nitrous oxide production rate. A higher nitrous oxide production was observed at a higher FNA concentration and nitrite concentration. Heterotrophic denitrification was put forward as the main production pathway.
- The inoculated biological air scrubber showed a faster start-up of nitrification and less nitrite accumulation during further operation. The inoculated air scrubber showed a higher ammonia removal efficiency and slightly lower nitrous oxide production than the non-inoculated air scrubber.

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Appendix 6A: Additional Figures

Table S6.1. Design specifications of the biological air scrubbers under study.

	Air scrubber 1 **3-stage crosscurrent**	**Air scrubber 2** **2-stage crosscurrent**
Dimensions(1) [m x m x m]	6 x 2.85 x 2.7 Stage1: 0.15 Stage2: 0.90 Stage3: 0.90	6 x 2.85 x 2.7 Stage1: 0.15 Stage2: 1.80
Packing volume [m^3]	15.0	15.0
Buffer tank volume (2) [m^3]	4.1 Stage1: 1.1 Stage2: 1.5 Stage3: 1.5	4.1 Stage1: 1.1 Stage2: 3.0
Specific surface area (2) [$m^2.m^{-3}$]	Stage1: 240 Stage2: 125 Stage3: 125	Stage1: 240 Stage2: 125
Maximum air flow rate [$m^3.h^{-1}$]	30720	30720
Water flow rate (2) Recirculation rate [$m^3.h^{-1}$]	Stage1: 20.0 Stage2: 16.5 Stage3: 19.0	Stage1: 19.5 Stage2: 19.5
Water distribution system (2)	Stage1: sprinklers Stage2 and 3: passive distribution gutter	Stage1: sprinklers Stage2: sprinklers
Minimal liquid to gas ratio [-]	0.0018	0.0012
Sprinkling density [$m^3.(m^2.h)^{-1}$]	10.0	7.0
Minimal EBRT [s]	1.76	1.76
Maximal LOAD [$m^3.(h.m^2)^{-1}$]	3992	3992

(1) Total length x width x height; Length of the three-stage and two-stage scrubber packing is given separately as well for each packing stage, with empty space between the stages.

(2) Dimensions for each scrubbing stage given separately.

Appendix 6B: Additional Figures

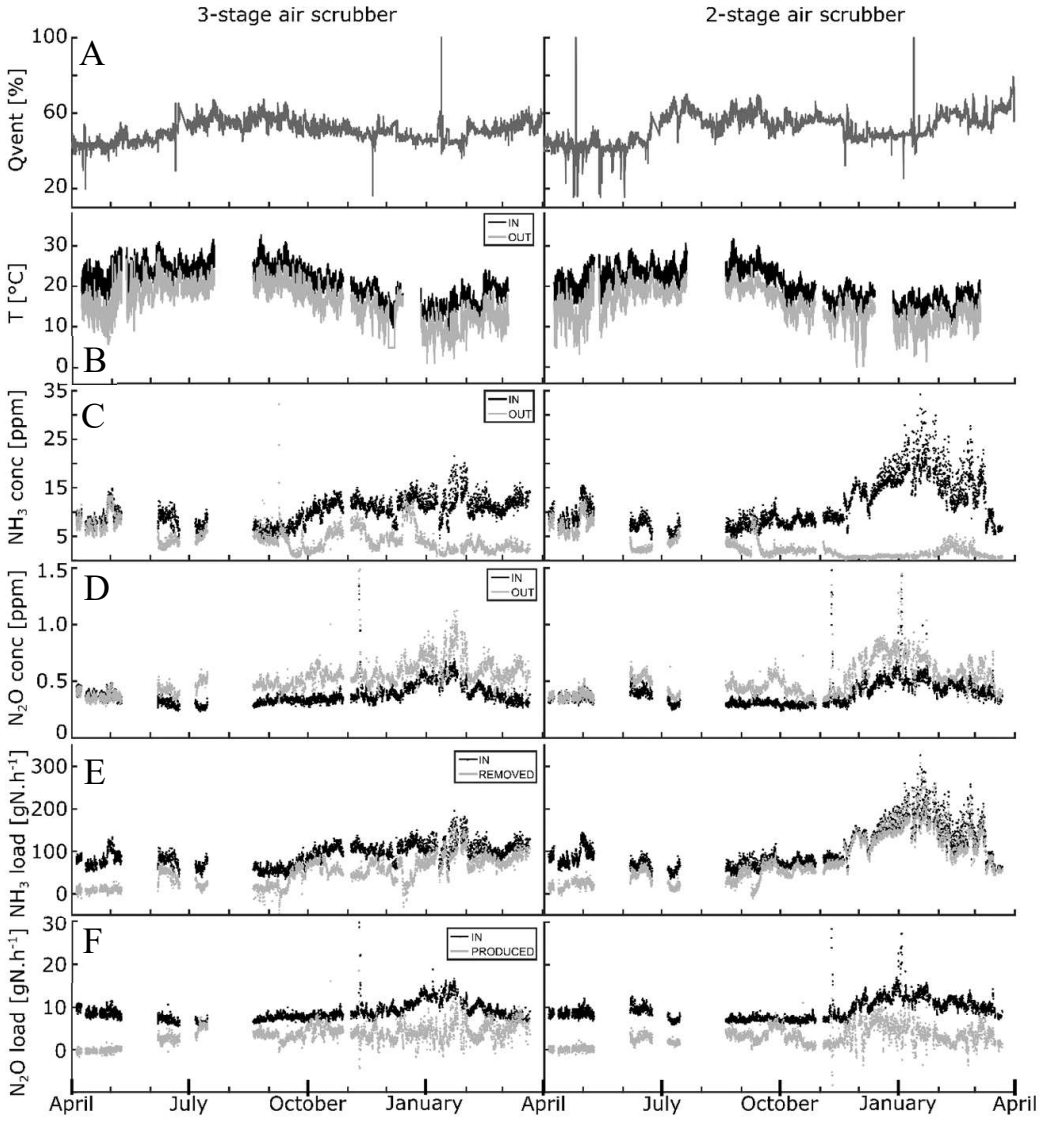

Figure S6.1 Overview of the ventilation rate, incoming and outgoing air temperature, ammonia and nitrous oxide concentration, as well as the ammonia and nitrous oxide loading rate and removal rate over the entire measuring period

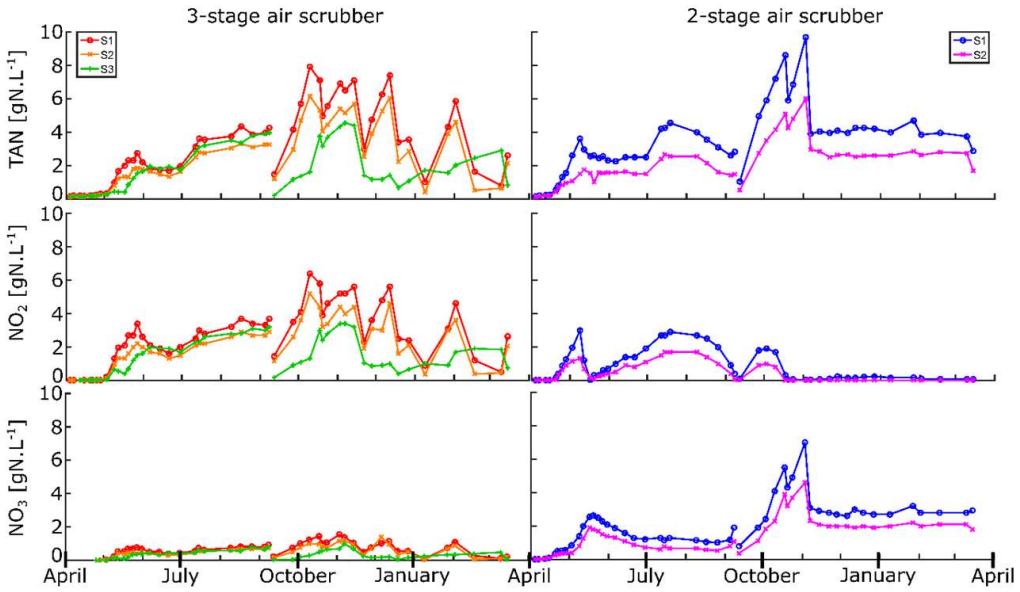

Figure S6.2 Overview of the total ammoniacal nitrogen (TAN), nitrite and nitrate concentrations in the washing water of the different buffer tanks over the entire measurement period.

Appendix 6C: Linear regression analysis

Table S6.2 Overview of the statistical linear regression analysis

		AS1 S1			AS1 S2			AS1 S3		
		p	**equation**	**R²**	**p**	**equation**	**R²**	**p**	**equation**	**R²**
NH3	**FA**	0.028	-0.4x+89.6	0.19	<0.001	-0.8x+94.6	0.43	<0.001	-1.3x+91.7	0.82
NO2	**FA**	0.015	-0.4x+76.9	0.23	0.010	-0.6x+73.3	0.26	0.007	-0.8x+65.9	0.30
NH3	**FNA**	0.365			0.600			0.511		
NO2	**FNA**	0.301			0.051			0.278		

		AS2 S1			AS2 S2		
		p	**equation**	**R²**	**p**	**equation**	**R²**
NH3	**FA**	0.110			0.002	-1.0x+92.6	0.36
NO2	**FA**	0.437			0.188		
NH3	**FNA**	0.049	-27.5+87.0	0.15	0.139		
NO2	**FNA**	0.142			0.007	88.2x+29.4	0.30

7

Long term measurement campaign at two full-scale biological air scrubbers: bacterial community structure

7.1 Abstract

In this study, the microbial community structure of two full-scale biological air scrubbers treating exhaust air from a pig housing facility were evaluated using 16S metabarcoding. The effect of inoculation with activated sludge of a nearby domestic wastewater treatment plant was investigated, which is a cheap procedure and easy to apply in practice. The study was performed at a three-stage and a two-stage full-scale biological air scrubber, of which only the latter was inoculated. Both air scrubbers evolved towards a rather similar community over time, which differed from the one in the activated sludge used for inoculation. However, the bacterial population at both air scrubbers showed small differences on the family level. A large population of heterotrophic bacteria, including denitrifying bacteria, was present in both air scrubbers. In the non-inoculated air scrubber, nitrite oxidizing bacteria (NOB) could not be detected, which corresponded with the incomplete nitrification leading to high nitrite accumulation observed in this system.

Van der Heyden, C., Demulder, T., Volcke, E.I.P., Demeyer, P., Heyndrickx, M., Rasschaert, G. (in preparation). Evaluation of the microbial population and dynamics at two full-scale biological air scrubbers at a pig housing facility – effect of inoculation.

7.2 Introduction

Pig production has intensified significantly during the last decades, resulting in potentially higher emissions of pollutants from pig housing facilities. Pig exhaust air contains a high nitrogen load, mostly as ammonia, and many odorous organic compounds, which are usually low in concentration but can have a significant impact on odour nuisance (BREF, 2017). Biofiltration of air with biological air scrubbers (or biotrickling filters) is frequently used as an effective technique for exhaust treatment of mechanically ventilated animal houses (Melse & Ogink, 2005). Intense contact between the polluted air and washing water over an inert packing material drives the water-soluble components from the gas phase to the liquid phase, thus cleaning the exhaust air.

Ammonia and other pollutants, like volatile organic compounds (VOCs), are taken up in the scrubber system and oxidized by bacteria. The microbial population of the biofilm is considered to be a consortium of nitrifiers and organoheterotrophs (Juhler et al., 2009). Nitrification mainly involves two phylogenetically unrelated groups of autotrophic bacteria, i.e. ammonia oxidizing bacteria (AOB), which convert the absorbed ammonium (NH_4^+) into nitrite (NO_2^-) and nitrite oxidizing bacteria (NOB), which convert nitrite further into nitrate (NO_3^-) (Ge et al., 2015).

Only a few studies on the microbial community in biological air scrubbers treating pig exhaust air are available (Blázquez et al., 2017; Juhler et al., 2009; Kristiansen, Lindholst, et al., 2011; Kristiansen, Pedersen, et al., 2011). It has been shown that heterotrophic bacteria comprise a large part of the biofilm and that nitrifying bacteria represent only a small fraction with less than 5% (Blázquez et al., 2017; Kristiansen, Pedersen, et al., 2011). Additionally, NOB often appear to be absent, resulting in nitrite accumulation (Juhler et al., 2009).

Inoculation of biological air scrubbers with activated sludge of a wastewater treatment plant is sometimes applied in practice in Flanders and the Netherlands. However, scientific results of the effects on microbial community are lacking.

In this study, the microbial community structure of two biological air scrubbers treating pig exhaust air was evaluated, additionally investigating the effect of inoculation on the microbial population. The study was performed with both a three-stage and a two-stage full-scale biological air scrubber system treating pig exhaust air. Only the two-stage air scrubber was inoculated with thickened activated sludge of a nearby domestic wastewater treatment plant. The bacterial population was monitored during the first three months of the start-up using 16S metabarcoding of samples collected at the different stages of the air scrubbers and in both the biofilm and washing water.

7.3 Materials and methods

7.3.1 Description of the air scrubbers

The study was conducted at two biological air scrubbers installed at one fattening pig housing facility, located in Flanders, Belgium. Air scrubbers 1 and 2 are a three-stage and a two-stage crosscurrent air scrubber, respectively (Figure 6.1). Both first stages, closest to the housing facility, are the dust sections with water being sprayed on top of the packing and from the front to remove as much dust from the exhaust air as possible. The second stages are called the bio sections. It is assumed that most ammonia is absorbed here. The third stage, only present at air scrubber 1, is generally referred to as the odour section as it is assumed that more odour treating bacteria can survive in this section. Electrical conductivity (EC) is monitored continuously with an EC sensor (ECDIND PT, Emec, Vazia, Italy) as a measure of total nitrogen in the washing water. At a threshold value of about 10 to 20 $mS.cm^{-1}$, corresponding to 2.3 to 5.1 $gN.L^{-1}$, the washing water is discharged from the first stage (dust section) into a tank. Fresh water is then added at the last stage of each air scrubber (bio or odour section respectively) to replace the discharged and evaporated washing water. The water thus flows in the opposite direction as that of the airflow, allowing the dirtiest washing water to come into contact with the dirtiest air and the cleanest water with the cleanest air, increasing the driving force of the latter. Consequently, the concentration of the pollutants increases in the washing water, from stage 3 to stage 1.

Both air scrubbers were started up in March 2016 and were fully operating in April 2016. Air scrubber 1 was not inoculated. Air scrubber 2 was inoculated twice at April 4th and September 12th 2016, with approximately 1 m^3 thickened activated sludge water from a nearby domestic wastewater facility in a buffer tank of 4 m³. Before the second inoculation, the last buffer tanks of both air scrubbers were emptied and refilled with fresh water. A continuous measurement campaign, measuring the performance of both air scrubbers with regard to ammonia removal and nitrous oxide production, was performed between April 2016 and March 2017. More details on scrubber set-up and the results of this measuring campaign can be found in Chapter 6. An overview of the performance and most important operational parameters are summarized in Table 7.1. Both air scrubbers performed well for ammonia removal after start-up, but the two-stage inoculated air scrubber showed the highest and most stable ammonia removal over time. The three-stage non-inoculated air scrubber suffered from incomplete nitrification characterized by nitrite accumulation.

Table 7.1. Overview the air scrubber performances and operational parameters (average values over period April 2016 till March 2017)

		Air scrubber 1 3-stage non-inoculated	Air scrubber 2 2-stage inoculated
NH_3 removal	[%]	50.51 ± 29.73	70.02 ± 27.43
N_2O production	[%]	39.1 ± 28.5	31.0 ± 23.9
pH	[-]	7.5 ± 0.6	7.1 ± 0.6
EC	[mS.cm^{-1}]	18.7 ± 13.0	20.6 ± 11.3
NH_4^+	[gN.L^{-1}]	2.5 ± 1.9	2.8 ± 1.8
NO_2^-	[gN.L^{-1}]	2.1 ± 1.5	0.6 ± 0.8
NO_3^-	[gN.L^{-1}]	0.5 ± 0.3	1.8 ± 1.3

7.3.2 Sample collection

Three types of samples were collected: from the activated sludge used as the inoculum, from the biofilm and from the washing water. At the two moments of inoculation, i.e. day 1 (April 4th) and day 162 (September 12th), activated sludge samples were taken. In the air scrubber, biofilm and washing water samples were taken in each stage at day 19 (April 22th), day 73 (June 15th),

day 185 (October 5th), and day 227 (November 16th). Composite biofilm samples were collected from various positions over the entire surface area of the packing. Washing water was sampled in the top layer (max 5 cm deep). All samples were collected in sterile 15 mL tubes and frozen at -20°C. An overview of all samples (including replicates) is given in Table 7.2.

Table 7.2. Overview of samples collected at different days and locations in the two biological air scrubbers. Colour codes correspond to the positions in the air scrubbers as in Figure 6.1.

	Air scrubber Stage					Code	Repli-cates	Sample		
	1 S1	1 S2	1 S3	2 S1	2 S2			Date	Day	Number
SL						SLUDGE	A - B	04/04	1	1 - 2
B						AS1-S2-B	A	04/22	19	3
W						AS1-S2-W	A	04/22	19	4
B						AS1-S3-B	A	04/22	19	5
W						AS1-S3-W	A	04/22	19	6
B						AS2-S2-B	A,B,C	04/22	19	7 - 8 - 16
W						AS2-S2-W	A	04/22	19	9
B						AS1-S2-B	A	06/15	73	10
W						AS1-S2-W	A	06/15	73	11
B						AS2-S2-B	A	06/15	73	12
W						AS2-S2-W	A	06/15	73	13
SL						SLUDGE	A - B	09/12	162	14 - 15
B						AS1-S1-B	A - B	10/05	185	17 - 18
W						AS1-S1-W	A - B	10/05	185	19 - 20
B						AS1-S2-B	A - B	10/05	185	21 - 22
W						AS1-S2-W	A - B	10/05	185	23 - 24
B						AS1-S3-B	A - B	10/05	185	25 - 26
W						AS1-S3-W	A - B	10/05	185	27 - 28
B						AS2-S1-B	A - B	10/05	185	29 - 30
W						AS2-S1-W	A - B	10/05	185	31 - 32
B						AS2-S2-B	A - B	10/05	185	33 - 34
W						AS2-S2-W	A - B	10/05	185	35 - 36
B						AS1-S2-B	A - B	11/16	227	37 - 38
W						AS1-S2-W	A - B	11/16	227	39 - 40
B						AS1-S3-B	A - B	11/16	227	41 - 42
W						AS1-S3-W	A - B	11/16	227	43 - 44
B						AS2-S2-B	A - B	11/16	227	45 - 46
W						AS2-S2-W	A - B	11/16	227	47 - 48

SL = Activated sludge sample
B = Biofilm sample
W = Washing water sample

7.3.3 DNA extraction and 16S Amplicon Sequencing

DNA extraction was carried out using the Powersoil DNA Isolation Kit (MOBIO Laboratories, Carlsbad, CA) according to the manufacturer's instructions, using 250 mg of biofilm, pellet from washing water or activated sludge. The washing water samples were centrifuged at 10 000 rpm for 1 min. The precipitate was scraped off and if not enough was present, remaining water was used to reach 250 mg sample. DNA quantity and quality was measured using the NanoDrop ND-1000 (Thermo Scientific, Wilmington, USA) and the Quantus double-stranded DNA assay (Promega, Madison, USA).

The taxonomic profiles of bacterial communities were determined using amplicon sequencing of the V3−V4 variable region of the 16S rRNA gene. The library preparation was based on the Illumina 16S metagenomic sequencing library preparation protocol (De Mulder et al., 2016; Illumina, 2013). The amplicon PCR was performed with the primers described by Klindworth et al. (2013). The final barcoded library was sequenced using Illumina MiSeq V3-technology (2 × 300 bp, paired-end) by Oklahoma Medical Research Foundation (Oklahoma city, USA).

7.3.4 Processing of the sequence reads

The amplicon sequencing dataset was demultiplexed by the sequencing provider and barcodes were clipped off the reads. The raw sequence data are stored in the NCBI Short Read Archive, accession number (xxxxxx). Primers were removed using Trimmomatic v0.32 (Bolger, Lohse, & Usadel, 2014). Different programs of the USEARCH software v7.0.1090 were used for the following steps, in combination with software packages PEAR and QIIME. Forward and reverse reads were merged using a minimum overlap length of 120 bp, a minimum and maximum resulting length of 400 and 450 bp and a quality threshold of 30 with a minimum length of 200 bp after trimming, using PEAR 0.9.8 (J. Zhang, Kobert, Flouri, & Stamatakis, 2014). The resulting sequences were quality filtered using "fastq_filter" with a maximum expected error of 3. Next, sequences of all samples to be compared were merged, dereplicated ("derep_fulllength") and sorted by abundance ("sortbysize").

UPARSE ("cluster_otus") was used for clustering the reads into operational taxonomic units (OTUs) at 97% identity level (Edgar, 2013). Chimeras were removed using UCHIME ("uchime_ref") with the RDP Gold database as a reference (Edgar, Haas, Clemente, Quince, & Knight, 2011). Finally, sequences of individual samples were mapped back to the representative OTUs using the "usearch_global" algorithm (97% identity) and converted to an OTU table using "biom convert" (McDonald et al., 2012). This procedure resulted in an average of 47581 sequences per sample with an average length of 416 bp (51 samples).

7.3.5 Downstream Data Analysis and Statistics

The composition and structure of the bacterial communities of the different sample types (activated sludge, biofilm, washing water) were analyzed. Rarefaction analysis were done using the R-package Vegan (Oksanen, G, & Kindt, 2015). Samples were retained if rarefraction curves indicated that the sequencing depth was sufficient (stationary phase was reached). The alpha-diversity was investigated by calculating the total number of observed species (rarefaction analysis) and estimating the diversity (Shannon−Wiener diversity index) using the Phyloseq package in R. For subsequent analysis of the betadiversity, only OTUs representing at least 0.01% of the total community in at least one sample were retained. Multivariate analysis of the dataset was done as previously described by De Tender et al. (2015), using the R package Vegan. The betadisper function was used to ascertain the multivariate spread of the data. Differences were visualised by constructing non-metric multidimensional scaling (nMDS) plots, using Bray−Curtis dissimilarity indices.

7.4 Results and discussion

7.4.1 Bacterial community structure

The bacterial community structure of the air scrubber samples was investigated. At 10 000 sequence counts, the rarefaction curves showed an average of 867, 279 and 198 different OTUs for the activated sludge, the

biofilm and the washing water samples, respectively (Figure S7.1). Sample number 5, 16 and 44 were not taken into account as not enough reads were found.

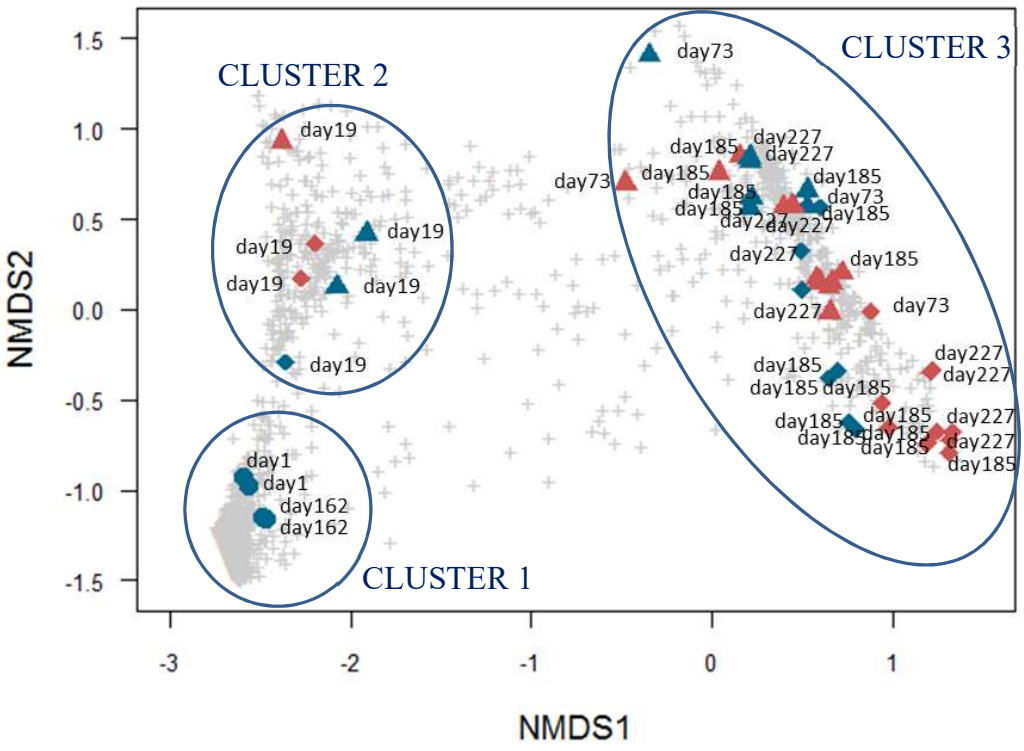

Figure 7.1. Non-Metric Multidimensional Scaling (nMDS) profile of pairwise community dissimilarity (Bray−Curtis) indices of 16S sequencing data of all samples. Shape represents different sampling type: ▲ biofilm, ♦ washing water and ● sludge. Samples of air scrubber 1 are indicated in red and of air scrubber 2 in blue. The sample date is indicated at each data point. OTUs are indicated in grey (+).

The non-Metric Multidimensional Scaling (nMDS) analysis visualises the differences in community composition between samples, taking into account the relative abundances of the species (Figure 7.1). The activated sludge samples clearly make up a separate clustering in the nMDS plot (cluster 1), indicating that these samples are different in richness and community composition compared to the biofilm and washing water samples. Two other main clusters were distinguished (cluster 2 and 3), which mainly differed through the effect of sampling day. Cluster 2 contained the samples from the first sampling at day 19 of both air scrubbers. Given their position on the plot, these samples were different from the activated sludge samples but still more related to the activated sludge samples than to the samples in cluster 3. Cluster

3 contained samples of biofilm and washing water of both air scrubbers at sampling day 73 till 227. Its position on the plot relative to the other two clusters indicated that the air scrubber bacterial communities evolve further away from those of the activated sludge samples. Cluster 3 comprised samples of both air scrubber 1 and 2, without a clear separation in the overall nMDS plot (Figure 7.1), although air scrubber 2 was inoculated whereas the air scrubber 2 was not.

If only the samples in cluster 3 are ordinated with nMDS, a more profound distinction, without any observed overlap, can be made between the two air scrubbers (Figure 7.2). The results hence show differences in the bacterial community composition between the two air scrubbers, but the differences between these two scrubbers are smaller compared to the difference between the two scrubbers and the activated sludge.

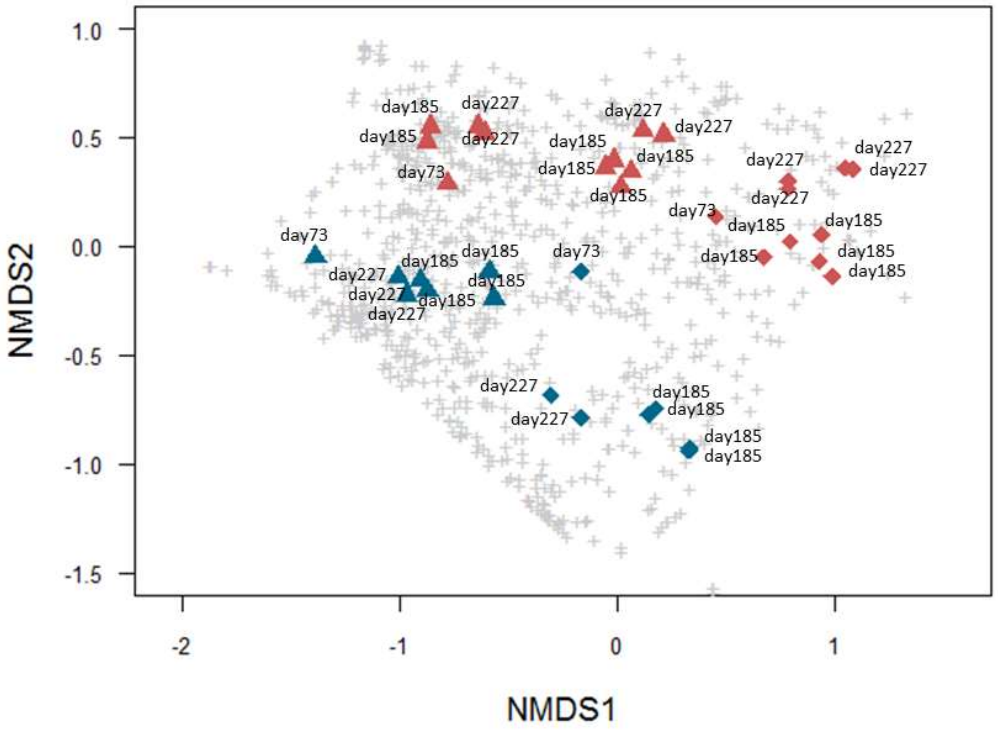

Figure 7.2. Non-Metric Multidimensional Scaling (nMDS) profile of pairwise community dissimilarity (Bray−Curtis) indices of 16S sequencing data of the samples of the last sampling period (cluster 3). Shape represents different sampling type: ▲ biofilm and ♦ washing water. Samples of air scrubber 1 are indicated in red and of air scrubber 2 in blue. The sample date is indicated at each data point. OTUs are indicated in grey (+).

The nMDS plots thus suggest that both air scrubbers, despite differences in configuration and inoculation, evolve towards a similar specialized community structure deviating from the activated sludge. However, when comparing only the results from the last sampling days, small differences in the bacterial community structure are observed, clustering the air scrubbers separately.

7.4.2 Composition of the bacterial communities

In the dataset, a total of 38 phyla and 342 families were identified across all samples. An overview of the bacterial community structure during the two last sampling days (185 and 227), when the population is considered to be in a dynamic steady-state, per air scrubber, stage and sample type, is given in Table 7.3. The overview of all samples on family level can be found in Appendix 7A (Figure S7.2). OTU's that could not be assigned to a specific taxon were grouped under "Unassigned". On average 1% of the OTUs could not be identified at family level.

The activated sludge samples showed a high relative abundancy of the phyla Proteobacteria (40%) and Bacteroidetes (29%). The other main phyla in the activated sludge samples were Actinobacteria (5.7%), Saccharibacteria (4.5%), Chloroflexi (5.0%) and Acidobacteria (3.6%) (Figure 7.3). The most abundant families found in the activated sludge samples were the *Comamonadaceae* (11%) belonging to the Proteobacteria and *Saprospiraceae* (11%), *Chitinophagaceae* (10%) and *Rhodocyclaceae* (5.0%) belonging to the Bacteroidetes. The phyla Proteobacteria, Bacteroidetes and Actinobacteria accounted for more than 80% of the community in all air scrubber samples. Chloroflexi and Acidobacteria represented less than 0.2% in the samples of the air scrubbers, showing a difference in community with the activated sludge samples. In addition, the family of *Rhodocyclaceae* (Proteobacteria) and *Saprospiraceae* (Bacteriodetes) were not very prevalent in the air scrubber samples compared to the activated sludge samples.

Table 7.3 Overview of the average relative abundance (%) and standard deviation of the major bacterial phyla and major families, discussed in this study. Sampling days 73 and 162 for the activated sludge samples and 185 and/or 227 for the air scrubber samples, were taken into account. A zero means that in none of those samples, OTUs of this phylum or family were found.

	Activated sludge	Air scrubber 1 – non-inoculated						Air scrubber 2 - inoculated			
Taxonomy		**Stage1**		**Stage 2**		**Stage 3**		**Stage 1**		**Stage 2**	
		Biofilm	**Water**	**Biofilm**	**Water**	**Biofilm**	**Water**	**Biofilm**	**Water**	**Biofilm**	**Water**
Sampling day	73 + 162	185	185	185 + 227	185 + 227	185 + 227	185 + 227	185	185	185 + 227	185 + 227
Proteobacteria	**40.1 ± 1.1**	**39.7 ± 0.5**	**50.8 ± 2.8**	**45.2 ± 2.8**	**51.2 ± 7.4**	**57.7 ± 8.8**	**38.0 ± 3.9**	**42.5 ± 0.4**	**24.7 ± 0.1**	**51.6 ± 2.2**	**26.3 ± 15.3**
Comamonadaceae	11.2 ± 5.1	17.0 ± 0.9	43.6 ± 4.3	22.8 ± 10.1	39.5 ± 3.5	27.9 ± 4.6	34.3 ± 4.6	14.4 ± 0.1	6.3 ± 0.5	14.8 ± 2.4	4.6 ± 1.0
Xanthomonadaceae	3,4 ± 0,8	5,5 ± 0.02	1,4 ± 0,3	6,7 ± 4,3	0,4 ± 0,2	14,2 ± 2.0	0,5 ± 0,2	14.0 ± 0,7	2,8 ± 0,2	17,9 ± 3,6	2,8 ± 2,8
Alcaligenaceae	0,4 ± 0,2	6,4 ± 1,4	2,4 ± 0,1	4,7 ± 2,9	2,2 ± 0,6	0,9 ± 0,3	0,5 ± 0,3	4,6 ± 0,2	6,8 ± 0,2	6,2 ± 3,3	8,7 ± 6,8
Rhodocyclaceae	5.0 ± 1.7	<0.1	<0.1	<0.1	0	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Nitrosomonadaceae	4,1 ± 0,1	0,8 ± 0.009	0,1 ± 0.02	1,5 ± 0,9	0,1 ± 0,1	10,7 ± 5,5	1,6 ± 1,1	0,6 ± 0,1	0,1 ± 0.04	1,2 ± 0,1	0,3 ± 0,3
Piscirickettsiaceae	<0.1	2,7 ± 0,7	1,4 ± 0,8	2,4 ± 3,2	3,7 ± 4.0	0,5 ± 0,3	0,1 ± 0.1	4.0 ± 0,5	3,6 ± 0,9	5,1 ± 1,5	4,5 ± 2,6
Methylophilaceae	<0.1	1,8 ± 0,3	0,4 ± 0.03	2,5 ± 1,6	2.0 ± 2,2	0,3 ± 0,2	<0.1	0,1 ± 0.01	0,1 ± 0.05	<0.1	<0.1
Bradyrhizobiaceae	0,6 ± 0,3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	1,2 ± 0,5	<0.1
Legionellaceae	<0.1	0.1 ± 0.002	0.2 ± 0.02	<0.1	0.2 ± 0.2	0.3 ± 0.2	0.2 ± 0.1	0.3 ± 0.1	<0.1	0.3 ± 0,3	0.1 ± 0,1
Bacteriodetes	**29.1 ± 0.5**	**45.0 ± 0.2**	**27.5 ± 1.1**	**38.4 ± 2.6**	**30.0 ± 3.7**	**21.6 ± 5.7**	**44.7 ± 2.4**	**33.0 ± 2.4**	**35.9 ± 0.5**	**29.0 ± 3.6**	**35.7 ± 10.1**
Flavobacteriaceae	0,6 ± 0,5	30,6 ± 0,2	16,2 ± 0,1	28,5 ± 2,5	20,2 ± 5,4	6,8 ± 4,8	32,9 ± 4,2	17,7 ± 1,9	7,5 ± 0,2	13,4 ± 5	8,9 ± 5,3
Cytophagaceae	0,2 ± 0,1	1,7 ± 0,1	6,6 ± 1,7	0,9 ± 0,9	6,1 ± 3,8	0,6 ± 0,3	10,4 ± 2,7	1,7 ± 0,5	20,9 ± 0,1	0,7 ± 0,5	15,6 ± 18,2
Chitinophagaceae	10,4 ± 4,1	9,2 ± 0,3	3,2 ± 0,3	5,4 ± 1,3	2,7 ± 0,3	7,8 ± 1,2	0,6 ± 0,2	9,3 ± 1,2	3,5 ± 0,5	9,5 ± 1,8	6,6 ± 3,3
Cryomorphaceae	0,1 ± 0,1	1,5 ± 0.05	0,5 ± 0.02	1,6 ± 0,6	0,6 ± 0,2	1,7 ± 0,9	0,4 ± 0,1	1,2 ± 0,3	3 ± 0,2	0,9 ± 0,7	3,4 ± 2.0
Saprospiraceae	11,2 ± 4.0	0,7 ± 0,2	0,1 ± 0.001	0,6 ± 0,2	0,1 ± 0,1	0,4 ± 0,2	<0.1	0,2 ± 0.03	0,1 ± 0.01	0,7 ± 0,5	0,1 ± 0,1
Actinobacteria	**5.7 ± 2.4**	**4.9 ± 0.01**	**12.2 ± 4.0**	**5.6 ± 2.1**	**8.4 ± 7.1**	**8.4 ± 2.1**	**6.9 ± 4.4**	**8.6 ± 1.8**	**22.8 ± 0.9**	**6.8 ± 3.4**	**20.6 ± 14.3**
Intrasporangiaceae	0,2 ± 0,1	1,4 ± 0.1	11,5 ± 3,8	1,8 ± 0,8	7,9 ± 7,4	2,5 ± 1.0	6,7 ± 4,4	3,2 ± 1,7	20,5 ± 1,3	3,5 ± 3,1	19,1 ± 14,5
Saccharibacteria	**4.5 ± 0.5**	**2.5** ± 0.2	**6.9** ± 0.5	**2.3** ± 0.9	**6.5** ± 4.7	**4.0** ± 1.1	**7.9** ± 3.8	**2.3** ± 0.1	**8.8** ± 0.2	**2.1** ± 0.4	**10.3** ± 2.5
Deinococcus-Thermus	**<0.1**	**2.4 ± 0.4**	**0.4 ± 0.2**	**4.1 ± 1.2**	**0.1 ± 0.1**	**1.2 ± 0.4**	**<0.1**	**5.4 ± 0.2**	**0.3 ± 0.1**	**2.9 ± 0.8**	**0.1 ± 0.2**
Trueperaceae	<0.1	2,4 ± 0,4	0,4 ± 0,3	4,1 ± 1,9	<0.1	1,2 ± 0,4	<0.1	5,7 ± 0,2	0,3 ± 0,1	2,9 ± 0,8	0,1 ± 0,2
Chloroflexi	**5.0 ± 0.7**	**0.1 ± 0.02**	**<0.1**	**0.1 ± 0.04**	**<0.1**	**0.1 ± 0.05**	**<0.1**	**0.2 ± 0.006**	**<0.1**	**0.2 ± 0.08**	**<0.1**
Acidobacteria	**3.6 ± 1.8**	**0**	**<0.1**	**<0.1**	**<0.1**	**0**	**0**	**<0.1**	**0**	**0.1 ± 0.1**	**<0.1**
Nitrospirae											
Nitrospiraceae	0.8 ± 0.3	0	0	0	0	0	0	<0.1	0	<0.1	0
Unassigned	**1.1 ± 0.3**	**1.9 ± 0.3**	**0.5 ± 0.03**	**1.3 ± 0.5**	**1.0 ± 0.8**	**1.8 ± 1.0**	**0.4 ± 0.3**	**1.8 ± 0.2**	**1.9 ± 0.2**	**1.7 ± 0.2**	**0.8 ± 0.2**

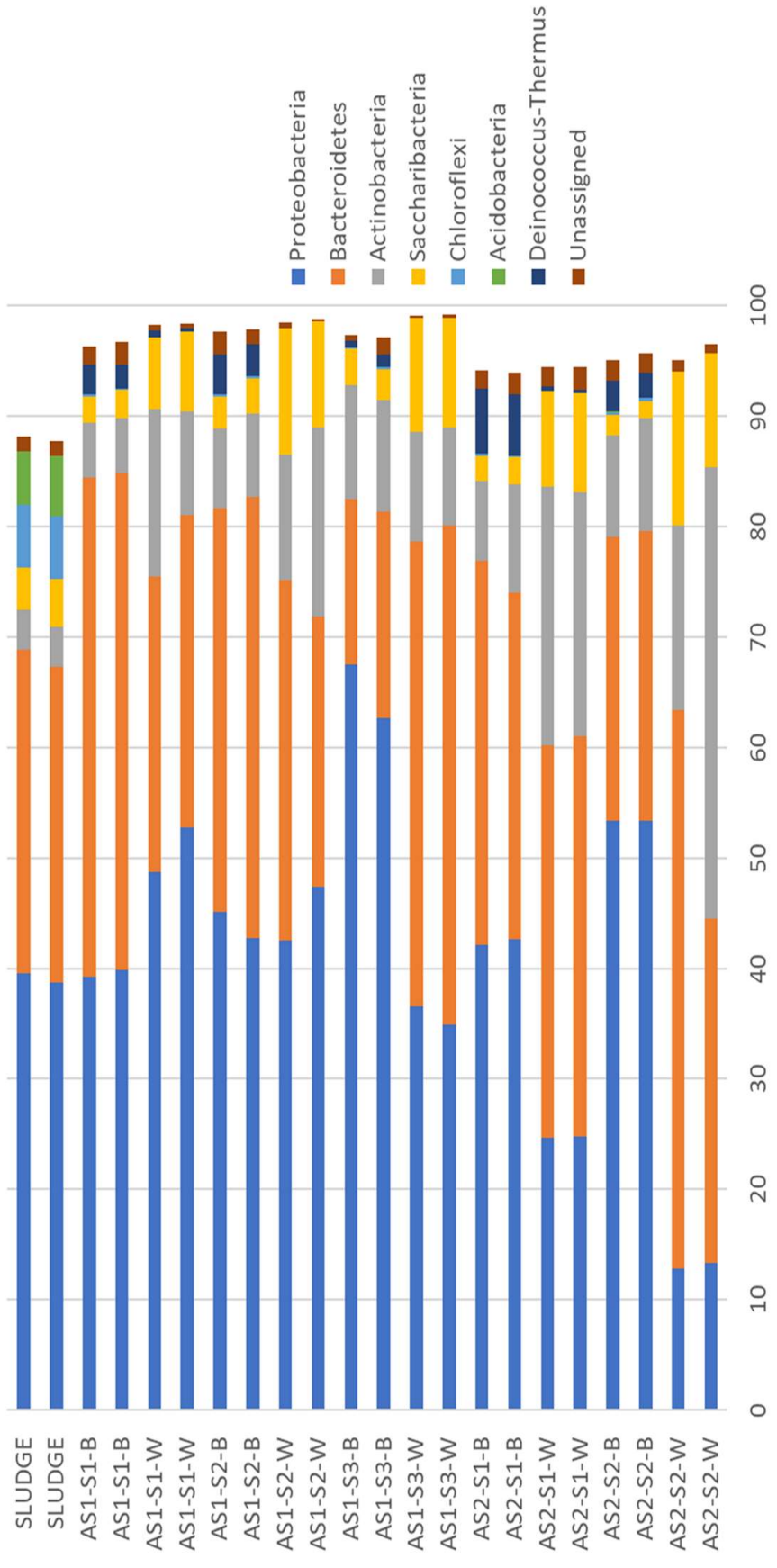

Figure 7.3. Bacterial phyla of the activated sludge sample compared to the bacterial profiles of the air scrubber samples, both in biofilm and water at day 185. Phyla that represent at least 2% in at least one sample are shown. OTUs that could not be assigned to a specific taxon were bundled in the group "Unassigned".

Previous studies on biotrickling filters and biofilters treating high levels of gaseous ammonia showed similar communities, with Proteobacteria, Bacteroidetes and Actinobacteria as the dominant phyla, although differing in relative abundance of the phyla (Blázquez et al., 2017; Kristiansen, Pedersen, et al., 2011). Most of the bacterial families found on air scrubber 1 were also found in air scrubber 2, but differences in relative abundances were found over the samples and between the air scrubbers.

One of the largest prevailing families in the air scrubber samples was *Comamonadaceae* (Proteobacteria), although its abundances differed from sample to sample (4.6% to 44%). In air scrubber 1, the abundance of *Comamonadaceae* was higher in the washing water samples, whereas in air scrubber 2, this family was found more in the biofilm samples This family was also abundantly present in other biological air scrubbers, treating pig exhaust air (Blázquez et al., 2017; Kristiansen, Lindholst, et al., 2011; Kristiansen, Pedersen, et al., 2011). It is a large and diverse bacterial family belonging to the order *Burkholderiales*, containing genera that includes aerobic organotrophs, anaerobic denitrifiers and Fe^{3+}-reducing bacteria, hydrogen oxidizers, photoautotrophic and photoheterotrophic bacteria, and even fermentative bacteria (Willems, 2014). Different genera belonging to this family were found in all samples. *Comamonas* is generally one of the most abundant microorganisms in biofilm communities driving wastewater treatment (Wu, Shukal, Mukherjee, & Cao, 2015). Under bulk aerobic condition, biofilms of *C. testosteroni* were capable of reducing nitrate, which even seemed to be beneficial for the biofilm lifestyle to reduce cell detachment (Wu et al., 2015).

As a consequence, *Comamonas* species can play a key role in denitrification under bulk aerobic conditions in biofilms (Blázquez et al., 2017). In our samples, the denitrifier C. nitrativorans was abundantly present, which has the ability to perform anoxic reduction of nitrate, nitrite and nitrous oxide to nitrogen (Etchebehere et al., 2001). Denitrification could explain why the N-balance in these two air scrubbers could not be closed (around 30% nitrogen loss; Chapter 6).

According to Kristiansen, Pedersen, et al. (2011), *Comamonadacea* are mainly responsible for the removal of highly soluble and easy degradable compounds as they are also known to utilize a variety of volatile fatty acids (VFA) and aromatic compounds under aerobic and denitrifying conditions. Juhler et al. (2009) also found that the aerobic activity was strongly dominated by heterotrophs, which accounted for 73 to 100% of the total oxygen uptake rate in the biofilm. It can thus be concluded that considerable amounts of heterotrophic families were thus present in the investigated air scrubbers.

The families *Flavobacteriaceae*, *Alcaligenaceae*, *Cytophagaceae*, *Cryomorphaceae*, *Piscirickettsiaceae* and *Trueperaceae* were present in some air scrubber samples, including the inoculated air scrubber 2, but were nearly not present in the activated sludge samples. The family of *Trueperaceae* is known to be capable of resisting great environmental hazards and to reside in wastewater (Griffiths & Gupta, 2007). It was furthermore found in high-strength ammonium wastewater, for example originating from landfill leachate (Miao et al., 2016; Tan et al., 2008). This could suggest more extreme prevailing conditions for bacteria in air scrubbers.

The family *Legionellaceae* was also present in some of the air scrubber samples at low abundancy (< 0.5%). The presence of the pathogen *Legionalla* was found in other studies (Blázquez et al., 2017; Kristiansen, Pedersen, et al., 2011) but not considered to be alarming (Melse, Schalk, & Bartels, 2015). Kristiansen, Pedersen, et al., (2011) even stated that at family level, the biofilm of a biological air scrubber treating pig house air contain a specialized bacterial community adapted to the unique extreme conditions in these biofilms, ensuring that microorganisms originating from pig feces, as well as pathogens, such as species of *Clostridium*, *Mycobacterium* or *Legionella*, are unable to establish in the biofilm due to a strong selective pressure.

Comparing the biofilm to the water samples, it appeared that in both sample types, the phyla Proteobacteria, Bacteroidetes and Actinobacteria were the most abundant. Air scrubber 2 showed a higher relative abundancy of Bacteroidetes and Actinobacteria, but a lower abundancy of Proteobacteria in the washing water samples compared to the biofilm samples. Additionally, a

clear difference in abundancy for the phylum Saccharibacteria was present between the biofilm and water samples, with a higher abundancy in the water samples. The biofilm samples had a higher abundance of the phyla Deinococcus-Thermus (3.3%). The latter was less than 0.2% represented in the water samples, and less than 0.1% in activated sludge sample, suggesting that this phylum has a higher preference for the biofilm environment. The families *Xanthomonadaceae* (Proteobacteria) and *Chitinophagaceae* (Bacteriodetes) were more prevailing in the biofilm and activated sludge samples, compared to the washing water samples. *Xanthomonadaceae* are abundant heterotrophs in water treatment systems that produce exopolymeric substances (EPS) involved in biofilm formation and *Chitinophagaceae* degrade a wide range of polysaccharides in biofilms (Dworkin, Falkow, & Rosenberg, 2006; Pal, Kraigher, Brajer-Humar, Levstek, & Mandic-Mulec, 2012). The families *Cytophagaceae* and *Intrasporangiaceae* were found more in the washing water samples than in the biofilm samples. Nevertheless, no taxonomic groups were unique for the biofilm or washing water environment. This could be expected as solid-adherent bacteria can end up in the washing water due to biofilm detachment.

When focusing on the difference in biofilm between stages of each air scrubber, it can be noted that the abundance of Proteobacteria increases from stage 1 to stage 2 and further to stage 3. This can be attributed to *Comamonadaceae* and *Xanthomonadaceae.* The pylum Bacteriodetes seems to decrease over the different stages. In the study of Kristiansen, Pedersen, et al. (2011) on a two-stage biological air scrubber with cellulose pads, a small difference between both stages was reported as well. In the first stage of that study, Bacteroidetes was the most dominant group, followed by Betaproteobacteria and Actinobacteria. In the second stage, Betaproteobacteria were most commonly observed, followed by Bacteriodetes and Gammaproteobacteria.

7.4.3 The nitrifying bacterial community

AOB are divided into two monophyletic lineages based on their respective 16S rRNA gene sequences (Junier et al., 2010). The first lineage belongs to the

Betaproteobacteria and comprises amongst others *Nitrosomonas* and *Nitrosospira* in the family *Nitrosomonadaceae* (Prosser, Head, & Stein, 2014). The second lineage, affiliated with the Gammaproteobacteria *in the family Chromatiaceae* (Klotz et al., 2006), contains *Nitrosococcus oceani* and *Nitrosococcus halophilus (Ward & O'Mullan, 2002) amongst others*. NOB are divided into four genera: *Nitrobacter* (family *Bradyrhizobiaceae;* de Souza, Alves, de Mello Varani, & de Macedo Lemos, 2014), *Nitrococcus* (family *Ectothiorhodospiraceae;* Imhoff, 2014), *Nitrospira* (family *Nitrospiraceae;* Daims, 2014) and *Nitrospina* (family *Nitrospinaceae;* Lücker & Daims, 2014) from which *Nitrobacter* and *Nitrospira* are the most important for nitrification (Ge et al., 2015). *Nitrospira* is regarded as K-strategist (with high substrate affinities and low maximum activity) for nitrite and oxygen, while *Nitrobacter* is an r-strategist under limited substrate conditions (Ge et al., 2015).

Focusing on the nitrifying community, in particular AOB, it was observed that the family *Nitrosomonadaceae* was present in all samples (Table 7.3). Almost all were represented by the genus *Nitrosomonas*, however, some traces of *Nitrosospira* were also found. *T*hese findings were in agreement with previous studies which reported *Nitrosomonadaceae* as the most prevailing family representing AOB in air scrubbers, and *Nitrosomonas* and *Nitrospira* as the most prevailing genera representing AOB in air scrubbers (Blázquez et al., 2017; Juhler et al., 2009; Kristiansen, Lindholst, et al., 2011; Kristiansen, Pedersen, et al., 2011). In the activated sludge samples, the abundancy of *Nitrosomonadaceae* was 4.1%. In the air scrubber samples, their abundance varied from 0.02% in the washing water up to more than 11% in some biofilm samples at both air scrubbers. At day 185 after start-up, a clear increase in abundance of *Nitrosomonadaceae* could be observed in the biofilm from stage 1 to stage 2 and stage 3 (if present). As a result of the overall countercurrent air-water flow in the air scrubbers, ammonia and VOC concentrations are expected to decrease from the air scrubber air inlet towards the outlet. Although generally the 2nd stage is referred to as the biological section for ammonia removal and the 3th stage the odour section for odorous component

removal, the results of this study show the opposite as more nitrifying bacteria are present at the last stages. This is in accordance with previous findings of Juhler et al. (2009) and Kristiansen, Pedersen, et al. (2011), which reported a higher abundancy of AOB at the 2nd stage compared to the 1st stage. Juhler et al. (2009) attributed this to a mixed biofilm community of heterotrophic and nitrifying bacteria competing for space and oxygen. Due to a significantly lower growth rate and dependency on oxygen, nitrifiers can only establish persistent populations in biofilm strata where the heterotrophs are limited by substrate and not by oxygen. As the exhaust air of pig housing facilities contains a large load of VOCs, heterotrophic activity is located mainly in the first stages and consequently, the nitrifying community will only be abundant at the last stages, where most soluble organics are not present anymore. The washing water at the last stages also contains the lowest pollutant load as fresh water is added there. Strikingly, removal of ammonia from the air is mainly accomplished by the first filter sections (Kristiansen, Lindholst, et al., 2011; Ottosen et al., 2011), where the nitrifying community is the lowest. It thus seems that absorption is sufficient to remove ammonia from the air, with the nitrifying population slowly oxidizing the absorbed ammonium in the water into nitrite and nitrate, without a considerable effect on the ammonia removal efficiency.

The NOB containing family *Nitrospiraceae* could be observed in some samples (Table 7.3). The family *Bradyrhizobiaceae* was present as well in some samples, but the genus *Nitrobacter* was not detected above the detection limit. Other studies showed no presence of any NOB (Kristiansen, Pedersen, et al., 2011), or only the genus *Nitrobacter* was present (Blázquez et al., 2017; Juhler et al., 2009). *Nitrospira* was present in the activated sludge samples at a low abundance of 0.80%. Interestingly, at the non-inoculated air scrubber 1, NOB could not be detected in any of the samples. The inoculated air scrubber, showed a low abundance of *Nitrospira* in the biofilm samples, but this differed over time. At day 19, 73, 185 and 227, the abundancy at stage 2 was respectively 0.3%, 0%, 0.0006% and 0%. This shows the low survival of NOB in air scrubbers and confirms that inoculation could help to introduce NOB in

the system, as NOB could establish together with the AOB during start-up as previously suggested by Juhler et al. (2009).

7.4.4 Inoculation and air scrubber performance

The presence of NOB in the inoculated air scrubber explains why it could establish full nitrification, without nitrite accumulation, while the non-inoculated air scrubber, showed a considerable nitrite accumulation (Table 7.1). High concentrations of nitrite are known to increase nitrous oxide production (Kampschreur et al., 2009). In Chapter 6, it was demonstrated that the inoculated air scrubber had on average a lower nitrous oxide production compared to the non-inoculated air scrubber (19%). As inoculation with activated sludge of a wastewater treatment plant is an easy and cheap method, it could thus be beneficial to inoculate biological air scrubbers in order to promote NOB, thus reducing nitrite accumulation and consequently lower the nitrous oxide production.

The nMDS plot (Figure 7.1) showed that the bacterial population of the inoculum evolves very quickly to an air scrubber specific microbial population, thus not surviving as whole. Activated sludge is a wide spectrum inoculum and thus contains a variety of bacterial families and species. In terms of ammonia removal and conversion in biological air scrubbers, only the phylogenetical groups AOB, NOB and (if present) denitrifiers, are important. A specific inoculum containing only AOB and NOB could therefore also be an option to apply, as in the study of Xue, Wang, Wu, Zhang, et al. (2010).

The removal of odorous components or less water-soluble components like methane, were in this study not taken into account. An optimized inoculum could in this perspective be beneficial. However, the operational conditions and the bacteria coming from the pig house exhaust air, contribute as well to which bacteria will survive and form a stable population. More research is necessary to investigate if an optimized inoculum would result in a better air scrubber performance. Additionally, this study was performed at two full-scale biological air scrubbers, during start-up. It must be further investigated if inoculation could result in a more stable NOB population at a nitrite

accumulating biological air scrubber in full operation.

7.5 Conclusions

- The microbial population at two full-scale biological air scrubbers treating pig housing outlet air evolved towards a similar specialized community structure, despite differences in configuration (3-stage versus 2-stage) and the fact that only one air scrubber was inoculated with activated sludge. However, when comparing only the results from the last sampling days, small differences in the bacterial community structure are observed. In both scrubbers, only a small percentage of nitrifying bacteria (AOB and NOB) and a large population of heterotrophic bacteria were present. The denitrifier Comamonas nitrativorans was abundantly present, confirming the possibility of denitrification in the prevailing bulk aerobic conditions.
- Different abundancies of some families were observed between the biofilm and washing water samples and between the different scrubber stages. Nevertheless, no unique taxonomic groups could be distinguished between the biofilm and the washing water environment. This could be expected as solid-adherent bacteria can end up in the washing water due to biofilm detachment. Nitrifying bacteria were more abundantly present at the last scrubber stages as they have problems of competing with the heterotrophs in the first stages.
- Inoculation with the wide spectrum inoculum activated sludge, which is a cheap procedure and easy to apply in practice, showed a positive effect on the air scrubber performance. In the inoculated air scrubber, NOB could be detected while this was not the case in the non-inoculated air scrubber. As a consequence, nitrite accumulation and nitrous oxide production was lower in the inoculated air scrubber, increasing the air scrubber performance.

Appendix 7A: Additional Figures

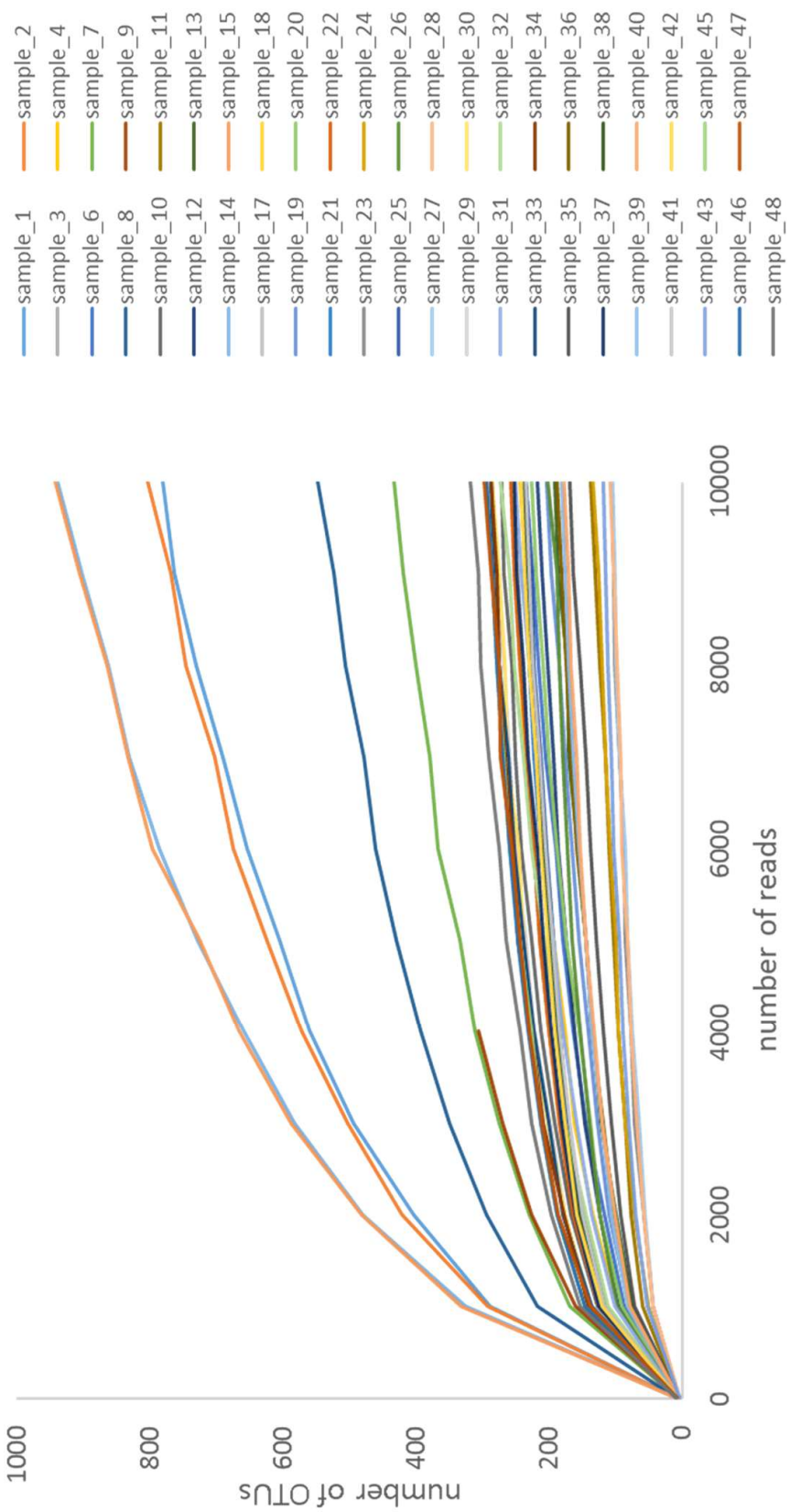

Figure S7.1. Rarefaction curve of bacterial communities in activated sludge, biofilm and washing water samples.

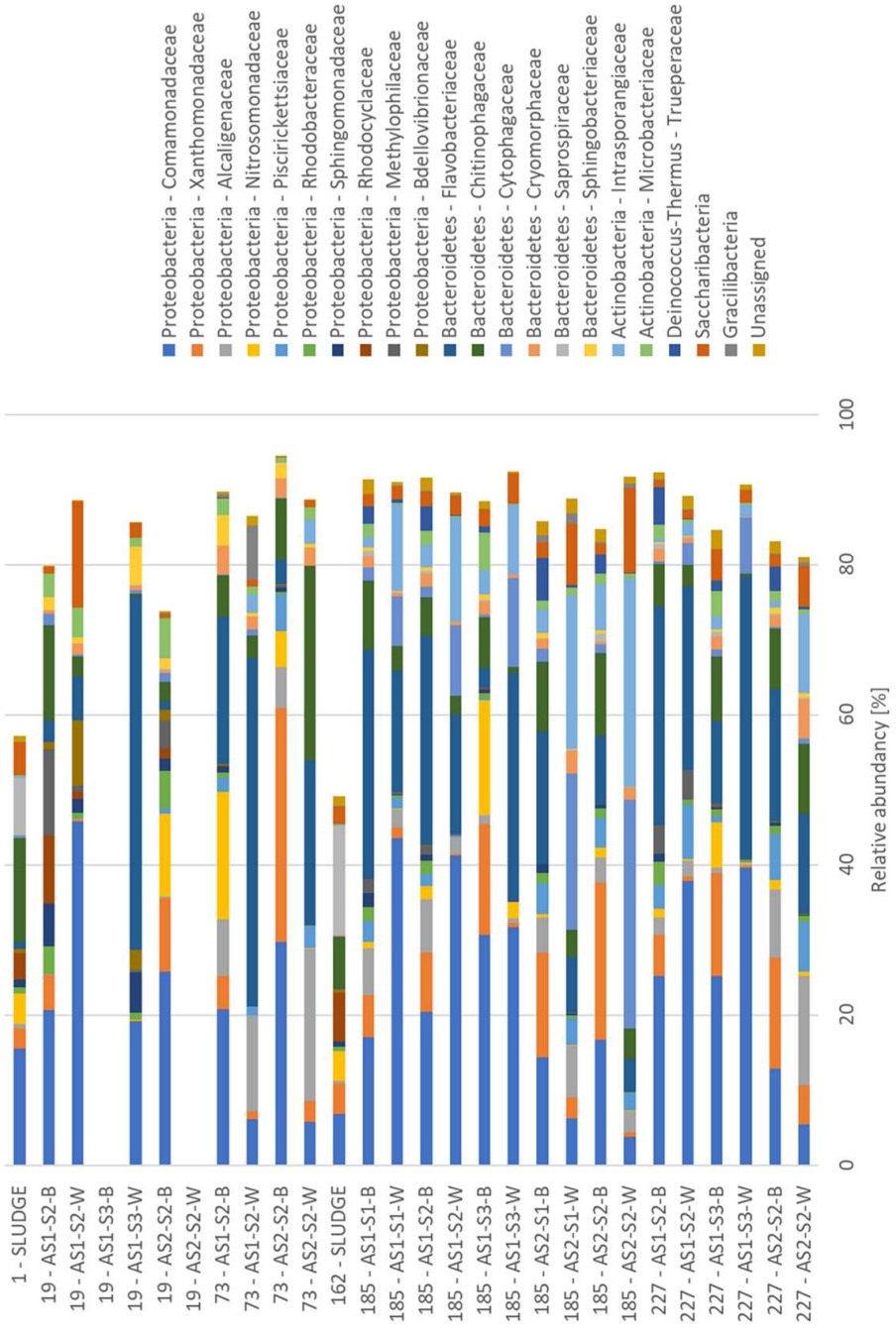

Figure S7.2 Bacterial family of all samples. Families that represent at least 5% in at least one sample are shown. OTUs that could not be assigned to a specific taxon were bundled in the group 'Unassigned'

8

General discussion, conclusions and perspectives

The **overall objective of this thesis** was to gain insight in the **design and operation of chemical, biological and multi-stage air scrubbers** for the removal of ammonia from pig housing facilities. Besides **ammonia removal** as the primary goal, attention was paid to the fate of the strong greenhouse gases **nitrous oxide and methane**, and to odour removal considering **hydrogen sulphide**. A combination of **mathematical modelling and simulation** (Chapter 4 and 5) and **full-scale experiments** (Chapter 3, 6 and 7) was applied. This chapter summarizes the key outcomes from this work, provides a general discussion and gives indications for future research.

Firstly, the use of air scrubbers as emission-low housing technique with legislation as a driver, is discussed (section 8.1). Next, an overview of the investment and operational costs is given (section 8.2) followed by a discussion on how to choose the appropriate system (section 8.3). Once the air scrubber is installed, the required removal efficiency needs to be reached, but it was shown that this is not always the case (section 8.4). The influence of air inlet conditions (section 8.5), design (section 8.6) and operation (section 8.7 to 8.9) on the scrubber performance was therefore assessed. Finally, attention was paid to the follow-up of air scrubbers performance during operation (section 8.10). This chapter is completed giving recommendations for practice and research perspectives.

8.1 Air scrubbers for ammonia emission-low animal housing – legislation as a driver

Building emission-low animal housing facilities, according to the European BREF, has become more important during the last decades to meet the increasingly stringent environmental legislation. Agriculture is responsible for the largest contribution of the anthropogenic emission of ammonia (UNECE/LRTAP, 2012), resulting in eutrophication and acidification. The first EU legislation to reduce ammonia emissions in agriculture was established in 1999 by the Gothenburg Protocol. With the implementation of the NEC directive (2001/81/EC), all member states, including Belgium and thereby the region Flanders, were forced to take actions. Since 2004, each

newly constructed pig and poultry housing facility has to be built according to an ammonia emission-low housing technique (MB19/03/2004), such as air scrubbers or source-based techniques. Despite 20 years of legislation and actions, **ammonia emissions from agriculture remains a hot item**. Flanders has many small, fragmented special protection areas identified as Natura 2000 sites that are surrounded by intensive agriculture. Negative effects arise due to the nitrogen input that exceeds the critical deposition load, leading to ecosystem degradation. Livestock farming in Flanders is facing a huge challenge in the context of the Programmatic Approach to Nitrogen (PAN) and is looking for practically applicable and affordable measures that can reduce the ammonia emission on farms in critical regions. **Odour nuisance** from animal housing facilities constitutes an additional problem in Flanders, where open space is limited and residential areas are often located next to the farms. With respect to **climate change**, the emission of the **greenhouse gases** such as nitrous oxide and methane, which are also present in the exhaust air from animal housing facilities, has received increased attention. The emission of **particulate matter** from agriculture has also gained more focus, given its effect on **human health** (ILVO/VILT, 2017). Overall, measures have to be taken to protect the environment and public health.

Air scrubbers are regarded as an appropriate solution to reduce emissions from animal housing facilities. At first instance, one may think that **front-of-pipe techniques**, avoiding the emission of components in the first place, should be preferred. However, even though being cost-effective, currently available front-of-pipe techniques, such as a reduced emitting surface area or adapted feed, do not always realize the high reductions required to meet strict permit regulations. Additionally, the effectiveness of these measures is mostly difficult to assess in practice. While air scrubbers are indeed **end-of-pipe solutions**, they do have the potential to achieve the **highest possible emission reductions**.

8.2 Costs inventory

In the following section, an estimation of the investment and operational costs of chemical and biological air scrubbers is given as well as a comparison with some front-of-pipe-techniques. Many different systems are on the market and the costs differ amongst others on design, capacity and manufacturer but also on the guaranteed removal efficiency. These numbers can thus only serve as an indication.

Investment costs of air scrubbers

An extensive analysis of the investments costs of German certified chemical and biological air scrubbers was made by KTBL (2008). Based on scrubbers designed for a capacity of 1000 to 1200 fattening pig places in 2005/2006, the investment costs varied between 47 to 72€ per fattening pig place. The investment costs of Dutch air scrubbers is estimated for 2017-2018 to vary between 30 and 34€ for chemical and 37 to 39€ per fattening pig place for biological air scrubbers (KWIN, 2017). The higher the achieved ammonia removal efficiency, the higher the investment cost. The prices are based on air scrubbers installed during the construction of a new animal housing facility. When air scrubbers are installed in existing animal houses, extra investment costs need to be considered. The building shell needs to be adapted to fit the air scrubber, such as the pressure chamber and the fans as they need to overcome higher pressure drops. Sometimes an extra storage for discharge water is needed or a safe acid storage and emergency shower in case of a chemical air scrubber. In some cases, more concrete is needed around the pig housing facility to be able to safely reach the air scrubber. These costs can drastically enlarge the investment costs.

Operational costs of air scrubbers

The most important operational costs of air scrubbers are the discharge of the wastewater, the consumption of fresh water, the electricity use and the use of acid in chemical air scrubbers. These costs were calculated as described below and summarized in Table 8.1 and Table 8.2. Costs are expressed per fattening pig place and per kg of removed ammonia, taking into account an emission

factor EF_{NH3} of 2.5 kg NH_3 per fattening pig per year and the ammonia removal efficiency η. The costs for control and maintenance, such as the working hours and the maintenance contracts were not taken into account.

The amount of discharge water (Q_{drain} [L.(fattening pig place.year)$^{-1}$]) depends on the ammonia loadthat is coming in and is being removed and the allowed total nitrogen concentration in the discharge water S^{max}_N. In chemical and biological air scrubbers, a maximal nitrogen concentration of approximately 30 gN.L^{-1} and 3 gN.L^{-1} is allowed, respectively. A price of 12€ per m³ discharge water was taken into account, considering transport and processing. The price to discharge manure in the Netherlands varies between 10 and 25€ per m^3 (KWIN, 2017). This can be lowered to around 3€ per m³ if the discharge water can be used on own land (Melse & Willers, 2004).

$$Q_{drain} = EF_{NH3} \cdot \frac{\eta}{100} \cdot \frac{1000 \times 14}{S_N^{\max} \times 17} \qquad \textbf{Eq.8.1}$$

The consumption of fresh water depends on both the amount of discharge water (Q_{drain}) and the amount of evaporated water (Q_{evap}). The latter depends on the water loss as well as on the applied ventilation rate The water loss through evaporation is on average 1.3 gH_2O.m^{-3} and is dependent on the temperature and moisture content of the incoming air (Melse & Willers, 2004). An average ventilation rate of 35 m³ per fattening pig per hour was assumed. A cost of 1€ per m³ fresh water was assumed.

$$Q_{evap} = \frac{S_{evap}}{\rho_{H2O}} \cdot Q_{air} \times 365 \times 24 \qquad \textbf{Eq.8.2}$$

The electricity use is mainly induced by the recirculation pumps and the power needed for the fans to overcome the extra pressure drop and is of course depending on the system. According to the study of Vrielink et al. (1997), the recirculation pumps used 47.4 kWh per fattening pig place per year and the fans 26.8 kWh per fattening pig place per year. For recently certified air scrubbers in Germany, the electricity use for the pumps varied between 15 and 61 kWh per fattening pig place per year, while the electricity use by the fans varied between 9 and 30 kWh per fattening pig place per year, depending on the system and the season (DLG6243, 2014; DLG6284, 2015). In this

calculation, an estimation of 50 kWh per fattening pig place per year was used and a cost of 0.08€ per kWh electricity was assumed.

Table 8.1. Overview of the operational costs for chemical air scrubbers

Operational cost	Cost		Chemical							
			70% removal				95% removal			
Water discharge	12	€.m^{-3}	48	L.(pig.y)$^{-1}$	0.58	€.(pig.y)$^{-1}$	65	L.(pig.y)$^{-1}$	0.78	€.(pig.y)$^{-1}$
			27.5	L.kg$_{NH3}^{-1}$	0.33	€.kg$_{NH3}^{-1}$	27.5	L.kg$_{NH3}^{-1}$	0.33	€.kg$_{NH3}^{-1}$
Water consumption evaporation	1	€.m^{-3}	399	L.(pig.y)$^{-1}$	0.40	€.(pig.y)$^{-1}$	399	L.(pig.y)$^{-1}$	0.40	€.(pig.y)$^{-1}$
			228	L.kg$_{NH3}^{-1}$	0.23	€.kg$_{NH3}^{-1}$	168	L.kg$_{NH3}^{-1}$	0.17	€.kg$_{NH3}^{-1}$
Water consumption discharge	1	€.m^{-3}	48	L.(pig.y)$^{-1}$	0.05	€.(pig.y)$^{-1}$	65	L.(pig.y)$^{-1}$	0.07	€.(pig.y)$^{-1}$
			27.5	L.kg$_{NH3}^{-1}$	0.03	€.kg$_{NH3}^{-1}$	27.5	L.kg$_{NH3}^{-1}$	0.03	€.kg$_{NH3}^{-1}$
Electricity	0.08	€.kWh^{-1}	50	kWh.(pig.y)$^{-1}$	4	€.(pig.y)$^{-1}$	50	kWh.(pig.y)$^{-1}$	4	€.(pig.y)$^{-1}$
			28.6	kWh.kg$_{NH3}^{-1}$	2.29	€.kg$_{NH3}^{-1}$	21	kWh.kg$_{NH3}^{-1}$	1.68	€.kg$_{NH3}^{-1}$
Acid consumption	0,60 (98% H_2SO_4)	€.L$_{acid}^{-1}$	2.9	L. (pig.y)$^{-1}$	1.71	€.(pig.y)$^{-1}$	3.9	L. (pig.y)$^{-1}$	2.33	€.(pig.y)$^{-1}$
			1.6	L$_{acid}$.kg$_{NH3}^{-1}$	0.96	€.kg$_{NH3}^{-1}$	1.6	L$_{acid}$.kg$_{NH3}^{-1}$	0.96	€.kg$_{NH3}^{-1}$
TOTAL					6.74	€.y^{-1}			7.58	€.y^{-1}
					3.84	€.kg$_{NH3}^{-1}$			3.17	€.kg$_{NH3}^{-1}$

Table 8.2. Overview of the operational costs for biological air scrubbers

Operational cost		Biological							
		70% removal				85% removal			
	Cost								
Water discharge	12 €.m^{-3}	480	L.(pig.y)$^{-1}$	5.76	€.(pig.y)$^{-1}$	583	L.(pig.y)$^{-1}$	7	€.(pig.y)$^{-1}$
		275	L.kg$_{NH3}^{-1}$	3.29	€.kg$_{NH3}^{-1}$	275	L.kg$_{NH3}^{-1}$	3.29	€.kg$_{NH3}^{-1}$
Water consumption evaporation	1 €.m^{-3}	399	L.(pig.y)$^{-1}$	0.40	€.(pig.y)$^{-1}$	399	L.(pig.y)$^{-1}$	0.40	€.(pig.y)$^{-1}$
		228	L.kg$_{NH3}^{-1}$	0.23	€.kg$_{NH3}^{-1}$	188	L.kg$_{NH3}^{-1}$	0.19	€.kg$_{NH3}^{-1}$
Water consumption discharge	1 €.m^{-3}	480	L.(pig.y)$^{-1}$	0.48	€.(pig.y)$^{-1}$	583	L.(pig.y)$^{-1}$	0.58	€.(pig.y)$^{-1}$
		275	L.kg$_{NH3}^{-1}$	0.28	€.kg$_{NH3}^{-1}$	275	L.kg$_{NH3}^{-1}$	0.28	€.kg$_{NH3}^{-1}$
Electricity	0.08 €.kWh^{-1}	50	kWh.(pig.y)$^{-1}$	4	€.(pig.y)$^{-1}$	50	kWh.(pig.y)$^{-1}$	4	€.(pig.y)$^{-1}$
		28.6	kWh.kg$_{NH3}^{-1}$	2.29	€.kg$_{NH3}^{-1}$	23.5	kWh.kg$_{NH3}^{-1}$	1.88	€.kg$_{NH3}^{-1}$
Acid consumption	0,60 €.L$_{acid}^{-1}$ (98% H_2SO_4)	n.a.		n.a.		n.a.		n.a.	
		n.a.		n.a.		n.a.		n.a.	
TOTAL				10.64	€.y^{-1}			11.98	€.y^{-1}
				6.09	€.kg$_{NH3}^{-1}$			5.64	€.kg$_{NH3}^{-1}$

For a chemical air scrubber, the amount of acid (Q_{acid}) dosed needs to be accounted for. It is assumed that the acid is only needed to compensate the pH increase caused by ammonia absorption, considering the stoichiometric molar ratio between H_2SO_4 consumption and ammonia removal R_{stoich} of 0.5 mole H_2SO_4.(mol NH_3)$^{-1}$. The density ρ_{H2SO4} was 1.84 kg.L^{-1} and the purity P_{acid} was 96%.

$$Q_{acid} = EF_{NH3} \cdot \frac{\eta}{100} \cdot R_{stoich} \cdot \frac{M_{H2SO4}}{M_{NH3}} \cdot \frac{1}{\rho_{H2SO4} \cdot P_{acid}} \qquad \textbf{Eq.8.3}$$

For chemical air scrubbers, electricity use and the applied sulphuric acid accounted for 59% and 25% of the operational costs, respectively (Table 8.1). For biological air scrubbers, the largest part (54%) of the operational costs were due to the discharge of water, while 38% was for the electricity consumption (Table 8.2). The operational costs for biological air scrubbers (10.64€ per fattening pig per year -70% ammonia removal) are significantly higher than for chemical air scrubbers (6.74€ per fattening pig per year -70% ammonia removal). These calculated costs are comparable with the situation in the Netherlands as the total yearly costs for chemical and biological air scrubbers, achieving 70% ammonia removal efficiency are 7€ and 11 € per fattening pig, respectively (KWIN, 2017). The higher costs for biological air scrubber is mainly because a lot more (ten times higher) discharge water is produced. This is because only a total nitrogen concentration of 3 gN.L^{-1} is allowed in the discharge water of biological air scrubbers compared to 30 gN.L^{-1} in chemical air scrubbers. Increasing the allowed total nitrogen concentration in the washing water of biological air scrubbers could reduce the operational costs.

Comparison with front-of-pipe techniques

The comparison can be made between air scrubbers (end-of-pipe technique) and front-of-pipe ammonia emission-low housing techniques. In the Netherlands, the investment cost for a manure pit with slanted walls and concrete slatted floors costs varies between 31 and 39€ per fattening pig per year (KWIN, 2017) and is thus comparable to the investment cost of air

scrubbers. However, the yearly variable costs are only 3 to 4.4€ per fattening pig per year.

8.3 Choosing the appropriate air scrubber system

Farmers must decide on which air scrubber to install at their housing facility as many different systems are available. In Flanders, each installed air scrubber must reach at least 70% ammonia removal efficiency. The first choice that has to be made is between a chemical and biological air scrubber. Both have advantages and disadvantages and different aspects such as the guaranteed removal efficiency, costs, safety and environmental impact can be taken into account.

Chemical air scrubbers are considered the most easy to control, thereby reaching ammonia removal efficiencies up to 95%, if well designed. However, less odour can be removed by these systems. Nitrous oxide and methane cannot be removed but at least are not produced. The results of a life cycle assessment (LCA) study showed that the acid scrubber had the lowest environmental impact compared to a biological air scrubber (De Vries & Melse, 2017). The main reason for this was the higher ammonia removal efficiency that can be reached. This implies that the ammonia removal efficiency is strongly determining the environmental impact. Chemical air scrubbers are cheaper in operational costs compared to biological air scrubbers as less discharge water is produced (section 8.2). However, the investment costs are mostly higher due to the extra storage tank for discharge water, which cannot be stored in the manure pit (risk of H_2S formation). The discharge water has a high fertilization value because the ammonium sulphate concentration is high but application to some fields can be hampered due to the high sulphate concentrations. Additionally, the discharge water is acid, concerning a risk for acidification of the soil. As the pH must remain below 4, regular follow up is needed to check whether the acid tank is not empty. Additionally, safety can be an issue. The use of 96% pure sulphuric acid at the farm, implies a risk for accidents (VILT, 2016). Sometimes salt formation can be detected at the exhaust air of chemical

air scrubbers. This was noticed during the short-term measurement campaign at a chemical air scrubber, as explained in Chapter 3.

Biological air scrubbers can also reach a high ammonia removal efficiency but because biological reactions are less easy to control, this is more difficult to guarantee. High odour removal is possible, but the risk for nitrous oxide production is also high. An LCA study showed that a high nitrous oxide production has a large effect on climate change (De Vries & Melse, 2017). Denitrification should not be used in biological air scrubbers as very high nitrous oxide productions were observed, up to 66% of the incoming ammonia (Melse & Mosquera, 2014). Biological air scrubbers are more expensive than chemical air scrubbers due to the larger amount of discharge water, which has a lower nitrogen concentration. The discharge water can be discharged in the manure pit, not needing an extra storage tank. Nitrite accumulation in biological air scrubbers can also form a risk for safety. Already several accidents happened whereby washing water or discharge water came into contact with the drinking water of animals, leading to animal death (VILT, 2017). This danger is less known than that of sulphuric acid and cannot easily be detected.

In the Netherlands, each produced air scrubber belongs to a certain air scrubber type, described in general by a technical leaflet (VROM, 2013). A fixed ammonia, odour and dust reduction potential is given to each air scrubber type, derived from measurements or expert knowledge. In Germany, each produced air scrubber system needs to be **certified** before installation in practice is allowed (DLG, 2016). In Flanders, air scrubbers must thus be properly designed to reach at least 70% ammonia removal efficiency, but they only need to comply with a general description of a chemical or biological air scrubber, respectively (MB31/05/2011). On the other hand, an air scrubber producer can bring an air scrubber on the market without much restrictions since no official measurements of the air scrubber performance are required. This means that **no guarantee** of proper functioning of the air scrubber is given to the farmers as such. To get a better view on the performances in practice, ILVO was asked

in 2016 by the Flemish government to carry out an extensive monitoring campaign at different air scrubber systems.

In 2010, Germany, Denmark and the Netherlands agreed to set up a standardized test-protocol to test and verify the environmental performance and operational stability of air cleaning technologies, within the framework of **VERA** (Verification of Environmental Technologies for Agricultural Production). The test-protocol is intended to provide reliable and comparable information on the performance of new technologies to farmers, authorities and other stakeholder, thereby stimulating the implementation of these technologies to meet the environmental challenges of livestock production within the EU (VERA, 2010). However, the protocol requires an extensive monitoring campaign at different farms and is thus **highly demanding and costly** in implementation. Additionally, it would require an independent certification organisation, such as DLG in Germany. In Flanders, this protocol is not yet adopted, considering its practical implications.

Moreover, only an ammonia removal efficiency of at least 70% is required in Flanders up to now. Some installed air scrubbers reach a higher efficiency, but this is not rewarded. Therefore, it was investigated if the air scrubber types described on the technical leaflets in the Netherlands and the certified air scrubbers in Germany, could be categorized in different ammonia removal efficiency ranges, e.g. 70% - 85% and 85% or higher based on the provided air scrubber design and operational characteristics, such as the EBRT, load, pH and EC (Van der Heyden, Zwertvaegher, Demeyer, & Brusselman, 2017). Such an approach would make the certification of air scrubbers for higher ammonia removal efficiencies, e.g. 85% or higher, much easier in Flanders. However, it was found in this study that there are **not well described air scrubber characteristics that can guarantee a certain reduction performance**. This is rather related to the combination of important parameters, together with the total design of the air scrubber by the manufacturer. Certification of each air scrubber separately, thus gives the most guarantee to a farmer that the air scrubber system is well designed and, if properly operated, can reach the certified removal efficiency.

8.4 Measuring air scrubber performance

Once an air scrubber is installed, the removal efficiency can be measured, to check if the required removal efficiency is actually reached. A proper operation of air scrubbers, reaching the required reductions, is essential for different **stakeholders**. The **Flemish authorities** need to make a correct estimate of the emissions in order to set-up an effective policy. **Society** has gained more policy participation and wants to be more informed. Air scrubber **manufacturers** and **farmers** need to be sure of the proper operation of their product and investment, respectively.

In Flanders, a **compendium method** is used to measure the momentary performance of air scrubbers in terms of ammonia removal efficiency (VITO, 2012). The first step in this procedure is to check whether the air scrubber performs spatially homogeneous. This is assured when the relative standard deviation (RSD) of the measured outgoing ammonia concentrations over 6 different sampling points is lower than 30% (Mosquera et al., 2014; VITO, 2012). In Chapter 3, the **homogeneity of the air scrubbers**, normally checked by gas detection tubes, was investigated in more detail using continuous data. In consequence, it was advised to check the homogeneity through the average standard deviation over the different measurement points. The interpretation of results expressed in terms of the relative standard deviation (RSD) is not straightforward since low average outgoing concentrations lead to high relative standard deviations, which could give the wrong impression that the air scrubber is not homogeneous.

Additionally, it was found during the various measurements at air scrubbers in this thesis, that caution should be paid to the possible **effect of wind** (results not shown in the previous chapters). Especially at cross flow air scrubbers placed in open fields, the wind can have a significant effect on the measurements of the outgoing concentrations from the air scrubber. At a strong wind and low ventilation rate, which occur regularly in winter periods, it was found with smoke test that wind entered the air scrubber and disturbed the emissions. These situations were easily observed by measuring CO_2 before and

after the air scrubber. When reductions in CO_2 are noted, this is due to wind dilution since carbon dioxide is not absorbed in the air scrubber. It could thus be considered at each ammonia removal efficiency measurement to use a CO_2 sensor before and after the air scrubber to guarantee a correct evaluation of the ammonia removal efficiency.

From literature and this doctoral research, it is clear that currently installed chemical, biological or combined air scrubbers do not always reach the required removal efficiencies for ammonia, odour and particulate matter (Chapter 1). Both the experimental (Chapter 3) and simulation (Chapter 4) results showed that nitrous oxide and methane were not removed in air scrubbers, due to their low water-solubility (high Henry coefficient). Nitrous oxide can even be produced in biological air scrubbers. In Chapter 3 and 6, it was found that **on average 2 to 5% of the incoming nitrogen was converted to nitrous oxide**, reaching even maximum values of 15%. This should of course be avoided as much as possible. Different design and operational conditions were investigated in this doctoral research to increase process understanding. The influence of inlet conditions (section 8.5) e.g. ventilation rate, air temperature, relative humidity and pollutant load was assessed. The design of air scrubbers (section 8.6) and process control, especially in terms of pH (section 8.7) and nitrogen components (section 8.8) was scrutinized, as well as the effect of inoculation of biological air scrubbers (section 8.9).

8.5 Influence of air inlet conditions

The exhaust air of pig and poultry housing facilities is characterized by its ventilation rate, temperature and relative humidity, besides its pollutant load (Chapter 1). These disturbance variables need to be considered in the air scrubber design; their dynamics can cause fluctuations in the performance of air scrubbers once in operation.

A long-term monitoring campaign at a biological air scrubber showed diurnal and seasonal fluctuations in the ammonia removal efficiency (Melse, Ploegaert, et al., 2012). The **diurnal variations** were confirmed in our short-term measurement campaign performed at three different air scrubbers

(Chapter 3). Melse, Ploegaert, et al. (2012) hypothesized that the fluctuating ammonia removal efficiency was mainly due to the increased temperature during the day, resulting in a lower solubility and thus negatively affecting the removal efficiency. However, an **increased outside air temperature also results in a higher ventilation rate, affecting the pollutant concentration by dilution and/or stripping and thereby the loading rate**. At a higher ventilation rate and constant emission factor, the incoming pollutant concentration in the air scrubber will be diluted. This was confirmed in the chemical air scrubber model (Chapter 4) and in practice for ammonia, nitrous oxide and methane for two air scrubbers (Chapter 3). An increased ventilation rate can also result in stripping of more pollutants from the manure, thereby increasing the incoming concentration and thus the loading rate, which was observed for ammonia at one air scrubber (Chapter 3). Additionally, the outgoing nitrous oxide concentration showed a small increase at increased ventilation rates but since the measurement was near the detection limit, it could not be concluded whether this was due to the increased ventilation rate or due to the noise on the measurement as such. It is thus impossible to test the effect of air temperature, ventilation rate and loading rate separately in practice as they are interrelated (Melse, Ploegaert, et al., 2012).

In this work, simulations with a mechanistic model were performed to test the individual effect of air temperature, ventilation rate and loading rate. It was shown for both a chemical air scrubber (Chapter 4) and a biological air scrubber (Chapter 5) that **rather the ventilation rate than the air temperature and load causes variations in the ammonia removal efficiency**. At a higher temperature, the effect of the decreased solubility is compensated by an increased mass transfer coefficient, having no significant overall effect on the removal efficiency. At a higher loading rate, the removal rate increased as well, resulting in a constant removal efficiency. At an increased ventilation rate, the ammonia removal efficiency decreased because of the decreased contact time. These simulation results indicate that **a lower ammonia removal efficiency is expected during daytime and in summer, when the ventilation rate is the highest**.

During the long-term measurement campaign (Chapter 6), the air scrubbers were switched off for maintenance for one day, while the ventilation was still on. On those moments, **significant stripping of ammonia, nitrous oxide and methane** was observed. This situation should thus be avoided as much as possible.

A widespread thought is that a higher incoming concentration from the housing facility will facilitate meeting the required reduction percentage of a pollutant. For this reason, emission-low housing techniques before an air scrubber are avoided. However, it was shown with the chemical and biological air scrubber model (Chapter 4 and 5), that a lower incoming ammonia concentration at constant ventilation rate results in a lower outgoing concentration and this does not influence the removal efficiency. This means that the emission, being the concentration multiplied by the ventilation rate ($gN.h^{-1}$) decreased. For the sake of environment and health, lower absolute emissions are more beneficial and thus lowering the incoming ammonia concentration can only be promoted. Applying a **required maximum emission instead of a removal efficiency** could in that perspective be a useful alternative in legislation. In Germany; a maximum emission instead of a removal efficiency for odour is already applied.

Concerning the operational costs, the mechanistic model of the chemical air scrubber, which include **temperature and relative humidity**, showed that the water consumption increased with increasing temperature and decreasing relative humidity of the incoming gas stream (e.g. in summer), while they **hardly affect the removal efficiency**.

8.6 Influence of process design

With respect to design, careful consideration should be given to packing dimensions, packing material, the air flow configuration and the number of stages.

Air scrubbers are in the first place designed for ammonia removal. The most important design parameters are the **retention time (empty bed residence**

time, EBRT) and the available contact surface area. The EBRT is determined by the incoming ventilation rate and the volume of the packing, the contact area is determined by the total amount of packing material and its specific surface area. For a constant ventilation rate, the ammonia removal efficiency increases with increased packing volume and specific surface area, as confirmed by model simulations for a chemical air scrubber (Chapter 4). Moreover, **realizing a packing volume increase by increasing the cross-sectional area (length or width) resulted in a smaller positive effect than increasing its height, for the same contact time and contact area**. This is explained by the decreased gas velocity associated with an increased cross-sectional area, reducing the mass transfer coefficient. Designing air scrubbers with a larger packing volume, seems to be beneficial for ammonia removal. For biological air scrubbers however, overdimensioning of the air scrubber should be avoided as this implies a higher risk for acidification of the system (see section 8.4).

More efficient removal of less soluble components like odorants and greenhouse gases, requires longer EBRTs, implying larger installations and thus higher costs. For odour these longer EBRTs are still possible to reach in air scrubbers, but for **methane**, an EBRT of several minutes is required, which seems **not realistic for applications at animal housing facilities**. New types of packing material, such as clay containing iron oxide, or even gravel to promote the growth of methane oxidizing bacteria, are promising for the removal of odour and methane but further research is needed to allow implementation. In any case, the extra costs because of larger installations to reduce methane in animal housing facilities with air scrubbers would by far exceed what can reasonably be asked from farmers.

Both **countercurrent** and **crosscurrent** air scrubbers are widely installed in practice. Countercurrent air scrubbers have the highest theoretical removal efficiency. Manufacturers also indicate their good self-cleaning capacity. However, a small dysfunction in the liquid flow nozzles would propagate further into the packing, involving a higher risk to get dry spots, while in a crosscurrent air scrubber, the horizontally flowing air will still get into contact

with other wetted areas. Additionally, countercurrent systems are mostly not easily accessible, hampering regular examination of the packing material and spray pattern. For this reason, the two newly installed full-scale biological air scrubbers for the long-term monitoring campaign were established as crosscurrent air scrubbers.

Another design aspect concerns the choice between **single-stage** or **multi-stage** air scrubbers. Multi-stage air scrubbers seem more reliable to reduce more pollutants and at higher efficiencies, since subsequent stages can take care of the pollutant load remaining from previous ones (Chapter 1). Additionally, water can be reused, reducing the discharge volumes. However, more stages can also imply a more complex process control. During the performed monitoring campaign at the two biological air scrubbers, keeping the EC setpoint in the different water tanks was found much more difficult for the three-stage than for the two-stage air scrubber (Chapter 6). Additionally, a concentration gradient will occur and the abundancy of different bacterial families will differ over the stages, as confirmed by the microbiological population analysis (Chapter 7). When dealing with a three-stage scrubber, the first stage is typically considered for fine dust removal, the second one as the biological section for ammonia removal and the 3th stage as the odour section for odorous component removal (Melse et al., 2008). However, this work showed that more nitrifying bacteria were present at the last stages. This was especially important for the AOB. In general, more heterotrophic bacteria than nitrifying bacteria were present in the air scrubbers, which is due to the high VOC load. This **concentration gradient** over the different stages can also have a negative effect. When the total ammonia load is low and everything is already absorbed in the first stages, ammonia absorption in the last stage is not high enough to compensate nitrification, resulting in acidification.

8.7 Influence of pH

In **chemical air scrubbers**, the pH is kept low (typically **pH < 4**) by adding acid to the washing water. The absorbed ammonia is then transformed to ammonium, thereby ensuring a high enough driving force for the transfer of

ammonia from gas to liquid to reach the required ammonia removal efficiency. This was confirmed by experimental measurements (Chapter 3) and through modelling and simulation (Chapter 4). During the measurement campaign, the pH in the chemical air scrubber was increased to 7 each time before washing water discharge, to ensure a neutral discharge product that could more easily be applied to the land. However, the ammonia removal efficiency immediately dropped to zero at that point. For environmental concerns, this procedure can thus not be allowed.

In **biological air scrubbers**, the pH must be kept **between 6.5 and 7.5**, according to the Flemish legislation. Absorption of ammonia from the gas phase to the liquid phase, increases the pH as one mole are protons per mole of ammonia absorbed are taken up to form ammonium. **Nitrification decreases the pH** as two moles protons are formed per mole of ammonia converted. The positive effect of nitrification **on the driving force for ammonia transfer** from the gas phase to the liquid phase was due to the induced pH decrease rather than by the reduction of the total ammoniacal nitrogen content as such (Chapter 5). As a result of the pH decrease associated with nitrification, **only about half of the absorbed ammonia is nitrified to nitrite and nitrate** in case no pH control is applied, which is most common. Further nitrification would result in a pH drop that inhibits the bacteria, thus preventing further conversion. The washing water of biological air scrubbers thus typically contains 50% ammonium to the total nitrogen content. This was observed at all measured biological air scrubbers (Chapter 3 and 6), and confirmed through simulations with the biological air scrubber model (Chapter 5).

For biological air scrubbers, **acidification of the washing water** is often observed in practice. This has been recently observed at the three-stage biological air scrubber studied in Chapter 6 (August 2017, results not shown). The last stage of the scrubber acidified as the pH dropped to 4.7. However, the ammonia removal efficiency amounted to more than 90%, with an outgoing ammonia concentration around 1 ppm. The absorbed ammonia was immediately transformed to ammonium at the low pH, increasing the driving

force between gas and liquid. The nitrous oxide production did not show a significant difference compared to the period without acidification, with an average production of 103% compared to the incoming nitrous oxide concentration. Additionally, the odour reduction was measured with olfactometry according to EN 13725, and amounted on average 42.0 ± 15.7%, based on 3 measurements (results not presented in the previous chapters). As stage 2 and 1 still had a pH of 6.6 and 6.8, respectively, the produced discharge water from stage 1 was not acid, limiting the risk for corrosion or H_2S production (Arogo, Zhang, Riskowski, & Day, 2000). More caution should be paid to this situation if only one stage is present. Additionally, a low pH is undesirable for nitrification, as it will result in higher inhibitive FNA concentrations in the washing water. A low pH further involves a risk for stripping HNO_2 (g) from the washing water, especially when high nitrite concentrations are present. From preliminary research results in the Netherlands, it appears that high concentrations of nitrogen oxides are measured in the exhaust air of biological air scrubber characterized by a low washing water pH (Melse, 2017). Why some air scrubbers acidify and how to solve this in practice is not yet completely understood and constitutes interesting item for further research.

It was shown by modelling that keeping the pH constant in biological air scrubbers affects the nitrogen component distribution in the washing water rather than the ammonia removal efficiency (Chapter 6). Without pH control, only half of the ammonia that is absorbed in the washing water can be oxidized by the biomass. When the pH is controlled at a constant value which is sufficiently low to allow ammonia mass transfer to the liquid phase but still high enough for bacterial growth, almost complete ammonium conversion can be achieved. However, the ammonia removal efficiency is hardly affected by this pH control, while operational costs for base dosing are substantial.

If complete ammonium conversion is achieved, it is clear that for each mole absorbed ammonia, one mole of base (NaOH) needs to be added to react with the net produced mole H^+ by nitrification. The amount of base needed (Q_{base}), can be calculated according to following equation:

$$Q_{base} = EF_{NH3} \cdot \frac{\eta}{100} \cdot R_{stoich} \cdot \frac{M_{NaOH}}{M_{NH3}} \cdot \frac{1}{\rho_{NaOH} \cdot P_{base}} \quad \textbf{Eq.8.4}$$

The density of NaOH is 2.13 $kg.L^{-1}$ and the purity P_{base} was assumed to be 50%. It can be assumed that 50% NaOH, costs as much as 98% H_2SO_4, being 0.6€ per L_{base}. The operation costs for biological air scrubbers increase to 12.96€ and 14.80€ per fattening pig per year for 70% and 85% ammonia removal efficiency, respectively (Table 8.3). This means an increase by around 23% compared to the normal operational costs (Table 8.2) and will thus drastically increase the operational costs of a biological air scrubber.

In Germany pH control systems are already applied in biological air scrubbers, dosing both acid and base when necessary to keep the pH between 6.5 and 7.5. When certifying these air scrubbers, the acid and base consumption is determined during an 8 week measurement campaign in summer and winter. For two biological air scrubbers installed at a fattening pig housing facility, the acid dosing was on average 0.8 and 1.6 L per fattening pig place per year and the base dosing was 2.3 and 0.05 L per fattening pig place per year (DLG6243, 2014; DLG6284, 2015). In one air scrubber, more acid than base was dosed while in the other the opposite occurred. During some measurement periods, neither acid or base needed to be supplied, meaning that the pH remained between 6.5 and 7.5. The base dosing is significantly less compared to the base necessary to achieve complete nitrification (4.69 L per fattening pig place per year; Table 8.3). Considering the negative effects of acidification, the application of a pH control system, adding both acid and base to keep pH within a relatively wide operating range at biological air scrubbers could therefore also be recommended in Flanders.

Table 8.3. Overview of the operational costs for chemical air scrubbers

Operational cost		Biological							
		70% removal				85% removal			
	Cost								
Water discharge	12 €.m^{-3}	480 275	L.(pig.y)$^{-1}$ L.kg$_{NH3}^{-1}$	5.76 3.29	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$	583 275	L.(pig.y)$^{-1}$ L.kg$_{NH3}^{-1}$	7.00 3.29	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$
Water consumption evaporation	1 €.m^{-3}	399 228	L.(pig.y)$^{-1}$ L.kg$_{NH3}^{-1}$	0.40 0.23	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$	399 188	L.(pig.y)$^{-1}$ L.kg$_{NH3}^{-1}$	0.40 0.19	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$
Water consumption discharge	1 €.m^{-3}	480 275	L.(pig.y)$^{-1}$ L.kg$_{NH3}^{-1}$	0.48 0.28	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$	583 275	L.(pig.y)$^{-1}$ L.kg$_{NH3}^{-1}$	0.58 0.28	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$
Electricity	0.08 €.kWh^{-1}	50 28.6	kWh.(pig.y)$^{-1}$ kWh.kg$_{NH3}^{-1}$	4.00 2.29	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$	50 23.5	kWh.(pig.y)$^{-1}$ kWh.kg$_{NH3}^{-1}$	4.00 1.88	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$
Acid consumption	0,60 €.L$_{acid}^{-1}$ (98% H_2SO_4)	n.a. n.a.		n.a. n.a.		n.a. n.a.		n.a. n.a.	
Base consumption	0.60 €.L$_{base}^{-1}$ (50% NaOH)	3.87 2.2	L.(pig.y)$^{-1}$ L.kg$_{NH3}^{-1}$	2.32 1.33	L.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$	4.69 2.2	L.(pig.y)$^{-1}$ L.kg$_{NH3}^{-1}$	2.82 1.33	L.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$
TOTAL				12.96 7.42	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$			14.80 6.97	€.(pig.y)$^{-1}$ €.kg$_{NH3}^{-1}$

8.8 Influence of nitrogen component concentrations

The total nitrogen concentration in the washing water is another important control variable in the operation of air scrubbers. The washing water must be discharged if the nitrogen concentration becomes too high, as this forms a risk of crystallization and the driving force between the gas and liquid phase cannot be guaranteed anymore. As measuring the total nitrogen content directly is considered too expensive for application at animal housing facilities, electrical conductivity (EC) or density electrodes are used instead. In Chapter 6, different **linear regressions** from literature **between EC and TN in biological air scrubbers** were compared with the one based on the long-term measurement campaign (Chapter 6). The correlation established in this study was also valid at higher TN concentration and was validated at extremer conditions. It can therefore be used to estimate the total nitrogen content at any biological air scrubber.

In a **chemical air scrubber**, the EC setpoint is mostly around 250 $mS.cm^{-1}$. This corresponds to a total nitrogen concentration which is about 3 times lower than the maximum **solubility of ammonium sulphate**, 60 $gN.L^{-1}$. Because of the low pH in chemical air scrubbers, all absorbed ammonia is directly converted into ammonium, allowing a high driving force. During the short-term measurement campaign (Chapter 3), **crystallization** of ammonium sulphate salts on the packing material and on the surroundings, was observed and thus caution must be paid in this respect.

For **biological air scrubbers**, the EC setpoint is mostly around 16 $mS.cm^{-1}$, corresponding to a total nitrogen concentration of around 3.2 $gN.L^{-1}$, the setpoint in Flanders (MB31/05/2011). It was shown in the short-term (chapter 3) and long-term monitoring campaign (chapter 6), that half of the total nitrogen remained present as total ammoniacal nitrogen, while the other half is oxidized to nitrite and/or nitrate. Thus by setting the nitrogen content, also the nitrogen components are set. FA in the washing water is of utmost priority as it determines the driving force between the gas and liquid phase. Additionally, free ammonia (FA) and free nitrous acid (FNA), are known to have an

inhibitory effect on both AOB and NOB. Through correlations, it was shown that FA had a negative impact on the ammonia removal efficiency but not on the nitrous oxide production. A higher nitrous oxide production was observed at a higher FNA concentration. As also denitrification seems a plausible process in biological air scrubber (chapter 6 and 7), the main production pathway is considered to be heterotrophic denitrification. Further research on these pathways is needed in view of reducing N_2O emission. This could be established by extending the biological air scrubber model with N_2O production pathways by nitrifiers (Pocquet et al., 2016) and denitrifiers (Hiatt & Grady, 2008) and by confronting the simulation results with experimental data.

During the long-term measurement campaign (Chapter 6), the free ammonia (FA) concentration varied between 0 and 145 $mg.L^{-1}$ and the free nitrous acid (FNA) concentration between 0 and 4.7 $mg.L^{-1}$. Even through the highest FA and FNA concentrations were higher than the inhibition levels reported by Anthonisen et al. (1976), the ammonia removal efficiency always remained above 70%. This indicates that the **biomass had adapted to the high concentrations** prevailing at air scrubbers. Further research is needed to confirm these findings for other installations.

8.9 Effect of inoculation

The effect of inoculation on the performance (Chapter 6) and the bacterial population community (Chapter 7) was studied by inoculating one of the two full-scale biological air scrubbers with activated sludge from a nearby waste water treatment system. A significantly **faster start-up of nitrification** was noted for the inoculated air scrubber, being 18 days faster than the non-inoculated air scrubber. It could not unambiguously be determined whether the long-term air scrubber performance was also improved through inoculation. The available data from the start-up did not show a significant difference in ammonia removal efficiency and nitrous oxide production between both air scrubbers. However, less nitrite accumulation was observed during further operation in the inoculated air scrubber, a higher ammonia removal efficiency

and slightly lower nitrous oxide production than the non-inoculated air scrubber was observed.

It was assumed that the inoculated air scrubber started faster with nitrification as AOB and NOB were present from the beginning. This was confirmed by the metagenomics study in Chapter 7. The inoculated air scrubber showed a higher abundancy of NOB, which were also found in the activated sludge samples. In the samples of the non-inoculated air scrubber, **NOB were not detected**, which corresponded with the incomplete nitrification and high nitrite accumulation observed in this system. The metagenomics study indicated as well that the two biological air scrubbers treating pig housing outlet air, displayed on the phylum level a rather comparable bacterial population. The three-stage non-inoculated air scrubber and the two-stage inoculated air scrubber evolved towards a rather similar community, which was compared to the activated sludge of a nearby wastewater treatment system, that served as inoculum. Differences in the bacterial community composition between the two air scrubbers existed, but the differences between these two scrubbers were smaller compared to the difference between the two scrubbers and the activated sludge. Activated sludge is a wide spectrum inoculum and thus contains a variety of bacterial families and species. In terms of ammonia removal and conversion in biological air scrubbers, only the phylogenetical groups AOB, NOB and if present, denitrifiers, are important. A specific inoculum containing only AOB and NOB could therefore also be an option to apply. Optimization of the inoculum to remove odorous components or less water-soluble components like methane, could in this perspective be beneficial. However, the operational conditions and the bacteria coming from the pig house exhaust air, contribute as well to which bacteria will survive and form a stable population. More research is necessary to investigate if an optimized inoculum would result in an even better air scrubber performance. Additionally, it must be further investigated if inoculation could result in a more stable NOB population at a nitrite accumulating biological air scrubber in full operation.

Although the ammonia removal efficiency was 12% higher and the nitrous oxide production was 19% lower in the inoculated air scrubber, absolute

comparison of both air scrubbers was difficult because they differ not only concerning inoculation but also regarding process configuration (two-stage compared to three-stage). Nevertheless, this study demonstrated the potential of inoculation at biological air scrubbers, as a cheap and easy-to-apply measure to **accelerate start-up and decrease the risk of nitrite accumulation**.

8.10 Follow up of air scrubber performance during operation

Once installed, proper operation of the air scrubber must be followed-up by the farmer. In Flanders, the farmer has to manually write down once a week the parameters enclosed in the instruction manual (MB31/05/2011). These include mostly pH, EC, volume of discharged water, volume of used fresh water and electricity use. In the Netherlands, it is mandatory since 2016 to have these parameters **electronically logged**. Additionally, the commitments of both producer and farmer must be made clear in advance. This allows a better follow-up of the system and provides more clarity if problems occur.

The **pH and EC sensors** which are permanently installed at the air scrubber and on which the wash water discharge is controlled, need to produce reliable measurements. In this thesis, it was observed during the monitoring campaign, that these sensors can show significantly different results compared to calibrated sensors in the lab. A more **regular calibration** of these sensors, e.g. each three months or even once a month, would be advisable. This is for example the case in Germany.

For the moment, in Flanders only the pH and EC are being followed-up and compared to the required values. However, it was shown that keeping these variables in between the desired ranges gives enough guarantee for proper operation of the air scrubbers. Exceeding these boundaries however, does not necessarily imply that the ammonia removal efficiency is not reached. It is therefore **more interesting to measure the ammonia removal efficiency directly**. This could be done rather easy and cheap by gas detection tubes or portable electrochemical sensors. However, continuous monitoring systems which could be permanently placed and logged at the air scrubbers would be

even more compelling. The use of **closed path laser-based optical measuring devices** was found interesting in the review of Chapter 1. Recently, a **new electrochemical sensor** was put on the market and tested in lab and field (Melse, Ploegaert, & Ogink, 2016; Mosquera et al., 2017). The device showed a high potential as continuous monitoring system of the ammonia removal efficiency at air scrubbers as it showed reliable results on long-term and it is relatively cheap and easy to handle. With such a system, follow-up of the air scrubbers would be much more straightforward, allowing faster action when the required removal efficiency is not reached.

Recommendations for practice

Different air scrubber systems are available on the market and it is difficult for a farmer to choose the appropriate system. Certification of air scrubbers, comparable to the situation in Germany, is not required at the moment in Flanders. Still, a certification system gives the most guarantee to a farmer that the air scrubber system is well designed and, if properly operated, is able to reach the certified removal efficiency.

Once installed, the follow up of the air scrubbers' performance is done by monitoring pH and EC in the washing water. For chemical air scrubbers, the pH must remain below 4 to guarantee a high ammonia removal efficiency at all times. In biological air scrubbers it was shown that keeping pH and EC in between the desired ranges gives enough guarantee for proper operation but exceeding these boundaries does not necessarily imply that the ammonia removal efficiency is not reached. It is therefore **more interesting to measure the ammonia removal efficiency directly**. Allowing a higher total nitrogen concentration than now applied in Flanders, will reduce the operational costs of biological air scrubbers.

For biological air scrubbers, a pH control system, applying acid and base when necessary, is recommendable. If the pH is too high, ammonia is not removed adequately as the driving force of mass transfer between gas and liquid decreases. When a biological air scrubber acidifies, the risk losing the nitrifying population exists, and also stripping of harmful HNO_2 can occur. Dosing acid and base when needed, will of course increase the operational costs. pH control systems are already applied in biological air scrubbers in Germany.

Inoculation of biological air scrubbers with activated sludge from a well working wastewater treatment plant is a cheap and easy method to apply in practice. It was shown that this leads to a faster start-up of nitrification, less nitrite accumulation, and it has a positive effect on the performance, decreasing N_2O production.

Research perspectives

Mechanistic models for both a chemical and biological air scrubber were set up, including mass and energy balances to simulate removal efficiencies, temperature profiles and the evaporation rate. These models can be further extended, validated or calibrated to increase process understanding and to be able to estimate performance more adequately. For the chemical air scrubber model, also the effect of ionic strength and salt formation could be taken into account. For the biological air scrubber model, the production pathways of nitrous oxide could be added, as well as the behaviour of more odorous components and methane. The biofilm and substrate concentration inside the biofilm could be modelled to increase knowledge about the heterotrophic growth in biological air scrubbers, as this was shown to be significant. Coupling the models with computation fluid dynamics (CFD) modelling would allow to simulate dynamic air streams, for detailed design purposes.

Why some air scrubbers acidify and how to solve this in practice is not yet completely understood and could be an interesting item for further research. Acidification can take place when the absorption of ammonia is not sufficiently high to compensate the pH drop by nitrification. This is more likely to happen if the biological air scrubber is over-dimensioned or in case the air scrubber consists of multiple stages. For a relatively low ammonia load, most ammonia is already absorbed in the first stages, resulting in acidification in the last stages. Additionally, it can be investigated if other incoming components can induce this acidification, e.g. acid odorous components or hydrogen sulphate. Biological H2S oxidation results in the formation of the strong acid H2SO4 (Y. Yang & Allen, 1994) and can lower to pH to 4 at incoming H2S concentration of around 5 ppm.

It was shown that inoculation at start-up of biological air scrubbers with activated sludge from wastewater treatment plants, leads to higher detectable abundancies of NOB, less nitrite accumulation and lower N2O emissions. It would be worth investigating if inoculation could also increase the performance of a nitrite accumulating biological air scrubber in full operation.

Further optimization of the inoculum to also remove odorous components or less water-soluble components like methane, could in this perspective be beneficial. More research is necessary to investigate if an optimized inoculum would result in an even better air scrubber performance.

In general, the working principle of air scrubbers used in livestock production is identical to that of those used in industry where high reductions can be achieved, especially for chemical air scrubbers. Simulations with both the chemical and biological air scrubber model showed that high reductions are possible (Chapter 4 and 5). However, both measurement campaigns (Chapter 3 and 6) showed that this is more difficult in practice. The question arises why air scrubbers have such difficulties of reaching the required removal efficiency. One of the explanations for the lower reductions in agriculture could be the highly varying and less controlled inlet conditions compared to many industrial applications. Especially the effect of the ventilation rate was shown to be significant. Additionally, it is reasonably to assume that dust, which includes many different components, has a potentially huge influence on the scrubber performance, not only in terms of clogging but also in biofilm and foam formation. The effect of dust, coming from e.g. different feed types, was not considered as such in this thesis but constitutes an interesting topic for further research. More knowledge is needed on the conditions leading to failure.

Another general challenge, particularly in agricultural applications, is that the installation and **operational costs for air scrubbers should be really low** to keep some economic margin, without compromising too much on the quality of construction and of monitoring and control systems. An even more significant difference with the industry sector is that **farmers follow-up the installation themselves**. While they are most knowledgeable in producing pigs and poultry, they are no process engineers. An air scrubber is mostly regarded as an extra burden for farmers. The consideration can thus be made, if it is not more advisable to also invest more research leading to tools for proper follow-up and maintenance of air scrubbers, as well as in knowledge dissemination, than to merely focus on optimization of design and control.

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Curriculum Vitae

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Basic education

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2007 – 2010
Faculty of Science, University of Antwerp
Bachelor in Applied Biological Sciences
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Extra training

UGent Doctoral Training Programme (Doctoral School of Bioscience Engineering)

8th Advanced Biofilm Course, University of Copenhagen, 7 – 12 October 2013

Metagenomics, from bench to data analysis, Earlham Institute Norwich, 19 – 23 September 2016

Employment

From 1 September 2012:
Ghent University, Department of Biosystems Engineering
Institute for Agriculture, Fisheries and Food Research (ILVO), Research Unit Technology and Food Science, Environmental technology

PhD Fellowship through UGent BOF starting grant prof. E. Volcke and ILVO scholarship (1/09/2012 - 31/12/2013)

PhD Fellowship through IWT scholarship (1/01/2014 - 31/12/2017)

- Research in view of obtaining a PhD degree:
  'Optimization of air scrubbers for the reduction of ammonia, odour and greenhouse gases from animal housing systems'
  Supervisor: Prof. dr. ir. Eveline Volcke and dr. ir. Peter Demeyer

- Supervision of master dissertation students (3)

Publications

Papers in international journals with reading committee

Hove, N., Demeyer, P., **Van der Heyden, C.**, Van Weyenberg, S., Van Langenhove, H.. (2016). Improving the precision of dynamic olfactometry according to EN 13725: a case study for pig odour. *Biosystems Engineering*, accepted (IF 2016: 2.044; Q1).

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Van der Heyden, C., Solon, K., De Keukeleire, L., Demeyer, P., Volcke, E.I.P. (in preparation). Model-based evaluation of ammonia removal in biological air scrubbers.

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Conference contributions

De Pue, D., **Van der Heyden, C**, Buysse, J. (2017) Air scrubbers as ammonia emission abatement strategy: beyond technical parameters. In: *Book of abstracts of 3th International Conference on Manure Management and Valorisation (ManuResource2017),* 27-28 November 2017, Eindhoven, the Netherlands (2 pages). *Oral presentation/Poster*.

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Van der Heyden, C., Volcke, E.I.P., Demeyer, P., Brusselman, E. (2015) Ammonia and greenhouse gas emissions from air scrubbers at pig housing facilities in Flanders. *Accepted for oral presentation at the 6th IWA conference on odours & air emissions (ODOUR2015),* 16-18 November 2015, Paris, France. Event was cancelled. In: *Book of abstracts of 2th International*

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Outreach to a wider audience

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Promotor: Prof. dr. ir. Kathy Steppe

BSc thesis

Van der Heyden, C. (2008). Vergelijkende levenscyclusanalyse (LCA) van een bio-ecologisch bouwmateriaal versus een klassiek bouwmateriaal met dezelfde functionele karakteristieken. Universiteit Antwerpen (in collaboration with VITO).
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Supervision of thesis students

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MSc in Bioscience Engineering: Agriculture, UGent

Title (Dutch): Methaanemissiereductie door pocketvergisting op melkveebedrijven: modelbouw- en simulatiestudie

2014-2015, Jolien Hoens, Julie Vandeput, Thomas Van Loo, Charlotte Versyck

Bachelor in Bioscience Engineering, UGent

Title (Dutch): Hernieuwbare energie op schaal van landbouwbedrijven – In the pocket?!

2012-2013, Bart Vanthillo

MSc in Bioscience Engineering: Agriculture, UGent

Title (Dutch): Modelbouw en simulatie van een chemische luchtwasser voor ammoniakverwijdering uit stallucht